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Title RADIOACTIVITY IN OCEANIC ORGANISMS

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Zinc-65 and chromium-51 (principally from the nuclear reactors at Hanford, Washington) and zirconium-95--niobium-95, ruthenium-103, and cerium-141 (principally from Russian fallout) were measured in marine organisms and particulate material taken off the Oregon coast. Gamma-ray spectrometry, carried out at Hanford Atomic Laboratories, was used in the analysis of almost one-hundred samples of organisms obtained from midwater trawl tows made at transects off Astoria, Newport, and Coos Bay, Oregon, between June 1961 and April 1962.

Euphausia pacifica was found to make up the largest portion of the biomass, showing up in sufficient quantity for radioanalysis throughout the year. 76% of the 220 trawls, each averaging 30 minutes at depth, gave sufficient euphausiids for radioanalysis (i.e. 40 g. or more per haul). If only nighttime hauls are considered, 82% of the tows provided sufficient euphausiids.

Zinc-65, formed by neutron activation in nuclear reactors

cooled by Columbia River water, provides a tracer of the river water at sea. Euphausiids near the mouth of the river contained up to 150 picocuries of zinc-65 per gram of dry weight, while even those taken 165 miles off Coos Bay (about 250 miles from the mouth of the river) contained about 10 picocuries/g, depending on season. Euphausiids taken off southern California and from Alaska had about an order of magnitude less zinc-65 (than Coos Bay), indicating the Columbia River to be a major source of this isotope, although a lesser amount of zinc-65 does appear to be due to fallout.

The Russian nuclear weapons tests of 1961 introduced a relatively heavy amount of fission products into the northeast Pacific Ocean. These were quickly taken up by the plankton so that the spectra of euphausiids, salps, and sergestids at times showed strong peaks due to zirconium-95 and niobium-95, ruthenium-103-106, and cerium-141. The maximum activity measured was 618 picocuries of Zr^{95}-Nb^{95} per gram of dry weight of euphausiids taken 45 miles off Astoria in November 1961. Many figures show the seasonal distribution of these fission products in euphausiids, indicating a "hotspot" of fallout off Astoria in November 1961 which gradually dissipated by the following spring. The figures show no apparent correlation between fallout radioactivity and zinc-65, probably indicating independent sources of the two.

Radioactivity at three trophic levels was examined using membrane filters to sample phytoplankton (first trophic level) and detritus. Euphausiids represent a second trophic level, while lantern fish (whose stomachs contained principally euphausiids) represent a third trophic level. Zinc-65 was found in the higher trophic levels, but appeared on filters only near the mouth of the Columbia River. The fission products (Zr^{95}-Nb^{95}, $Ru^{103-106}$, and Ce^{141}) appeared both on the filters and in euphausiids, but were greatly discriminated against at the third trophic level. Chromium-51, formed at Hanford by neutron activation, was the most prominent gamma emitter on filter samples taken near the mouth of the river, but was nearly absent at higher trophic levels.

Euphausiids are considered to be important agents in the vertical transfer of radionuclides in the ocean because they: (1) concentrate radioactivity, (2) make up the largest biomass in the macroplankton, (3) are forage for many predators; and (4) make extensive vertical migrations across the thermocline, feeding at the surface at night, and moving into the depths in the daytime.

RADIOACTIVITY IN OCEANIC ORGANISMS

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I INTRODUCTION

One of the events observed after the 1956 atomic weapons tests in the central Pacific was the great affinity of marine plankton for radioactivity (42, p. 7). Relative to sea water, the concentration of the activity in the plankton was very great; often so great that animals high in the food web, such as tuna, became sufficiently radioactive as to be of doubtful value for human consumption (36, p. 861).

In addition to the sporadic, massive infusions of radioactivity into the biosphere due to atomic weapons tests, there are secondary low level sources of contamination attributable to nuclear reactors. The reactors used at Hanford, Washington, for the production of plutonium are responsible for perhaps the greatest single, continuous flow of radionuclides into the marine environment in the United States. The radionuclides are carried to the sea by the Columbia River, which because of its size, dilutes the effluent sufficiently to permit the continued use of the river for domestic purposes. Nevertheless, certain organisms in the river are known to accumulate radioactivity (21), and marine organisms would likewise be expected to concentrate some of the radioactive elements in the effluent. The objectives of this study were, first, to determine if measurable amounts of radionuclides from the river

were present in oceanic species and whether any specific organisms could be used routinely as biological monitors of the radioactivity at sea.

Because of the moratorium on nuclear testing, fallout was at a low ebb when this research was begun. In the absence of fallout, the radioactivity of marine organisms from this area near the Oregon coast might be expected to be closely associated with the Columbia River radionuclides. At least it might be assumed that the bulk of the short half-lived non-fission radioisotopes, such as Zn^{65} or Cr^{51}, were produced at the Hanford reactors. There was no assurance in the literature, however, that any radioisotopes could be found in oceanic organisms in the absence of nuclear testing, except directly at the mouth of the Columbia River.

The Russian nuclear tests of September 1961, which marked the end of the moratorium, quickly made their influence felt in the region off the Oregon coast. This unexpected source of radioactivity greatly exceeded the total radioactivity of the river effluent and tended to confuse the radioactivity pattern temporarily. However, it soon was discovered that the effects of the river were never completely masked by fallout because the two sources contributed different gamma emitters. Those from the Columbia River were formed principally by neutron activation, while the Russian fallout contributed mostly fission products.

With the advent of Russian testing, this study was extended to include the concentration of fallout radionuclides in macroplankton, and the vertical and horizontal transport of these radionuclides, as well as neutron-induced Zn^{65} from the river, by plankton. Finally, an effort was made to trace both biologically important and biologically unimportant radioisotopes through various trophic levels of the food web. Organisms chosen were those which appeared in quantity in midwater trawls made at a standard series of stations off Astoria, Newport, and Coos Bay, Oregon.

II RADIONUCLIDE SPECIES INVOLVED

A. Neutron-Induced Isotopes (from the Columbia River)

In 1944, three nuclear reactors for the production of plutonium were placed in operation at Hanford. Since then, five more have been added. These eight production reactors are cooled by the circulation of water from the Columbia River, which after a brief delay, is returned to the river. The coolant water is subjected to an intense neutron flux in the reactor, and these neutrons enter into the nuclei of many of the trace elements present in the water. Often the isotope formed is unstable because of the increased neutron-proton ratio, and decays radioactively, according to its nuclear species. These neutron-activation

products make up the majority of the radioisotopes found in the Columbia River. A few fission products are found (from natural uranium in the water, for example), but they occur in relatively small quantities (48, p. 8).

Some twenty radionuclides make up 98 percent of the radioactive material present in the reactor effluent. The balance (two percent) consists of forty other radioactive elements (48, p. 9). In addition to the radionuclides in the effluent, some volatile and particulate radioactive materials escape into the air from the 200-foot stacks, although about 99 percent of these are trapped and removed (48, p. 23).

In the vicinity of Hanford, the following seven radionuclides are encountered in the water, which make up 90 percent of the radioactive material four hours after irradiation in the reactors (48, p. 9):

RADIONUCLIDE	HALF-LIFE
Mn^{56}	2.58 hrs.
Cu^{64}	12.8 hrs.
Na^{24}	15.1 hrs.
As^{76}	26.8 hrs.
Si^{31}	2.62 hrs.
Cr^{51}	27.8 days
Np^{239}	2.33 days

Only the last two, Cr^{51} and Np^{239}, reach the ocean in quantity. The other five radionuclides do not reach the ocean in measurable amounts because:

1. They have short half-lives; since the average length of time for any portion of the river water to travel from Hanford to the ocean is about two weeks, a large percentage of the radioactive materials will have decayed enroute.
2. Some of the biologically important radionuclides are actively accumulated by organisms in the river (21). Although eventually these nuclides will be freed from the organisms and may return to the river, the delay may be great enough to cause the virtual disappearance of the nuclide by radioactive decay.
3. It is known that deposits of silt containing significant quantities of radioisotopes are accumulating in some sections of the river (54). While erosion will cause much of this material to eventually reach the ocean, it will have lost some of its radioactivity during the delay.

The determination of the total amount of any radioisotope reaching the ocean is difficult, even though the amount being released at Hanford may be known accurately. The difficulty arises because of the complex nature of the estuarine environment between Hanford and the ocean.

Tides bring oceanic water in and out. The fresh water from the river, whose pH may vary with the amount of acids or alkalis it contains, mixes with the buffered sea water, which is alkaline. Substances whose solubility is affected by pH (such as Fe ions) are precipitated. Also, there may be a "salt effect" on colloidal material, caused by increased ionic strength (due to the Na and Cl ions in the sea water). Turbulence caused by the interaction of the river and tides may cause an increase in particulate matter in suspension on which the radioisotopes can be adsorbed. Lastly, the oceanic water is difficult to analyze for radioactivity because it contains so much salt.

For these reasons, calculations on the amounts of radionuclides which reach the ocean from Hanford are based on the determinations of amounts of these radionuclides which pass Vancouver, Washington, on an average day (48, p. 22). Vancouver is above the estuarine stretch of the Columbia River (Figure 1). According to this evaluation scheme, the four most important radionuclides from Hanford in the Pacific Ocean, and their rates of flow past Vancouver, are (48, p. 22):

RADIONUCLIDE	CURIES PER DAY
P^{32}	17
Cr^{51}	850
Zn^{65}	38
Np^{239}	72

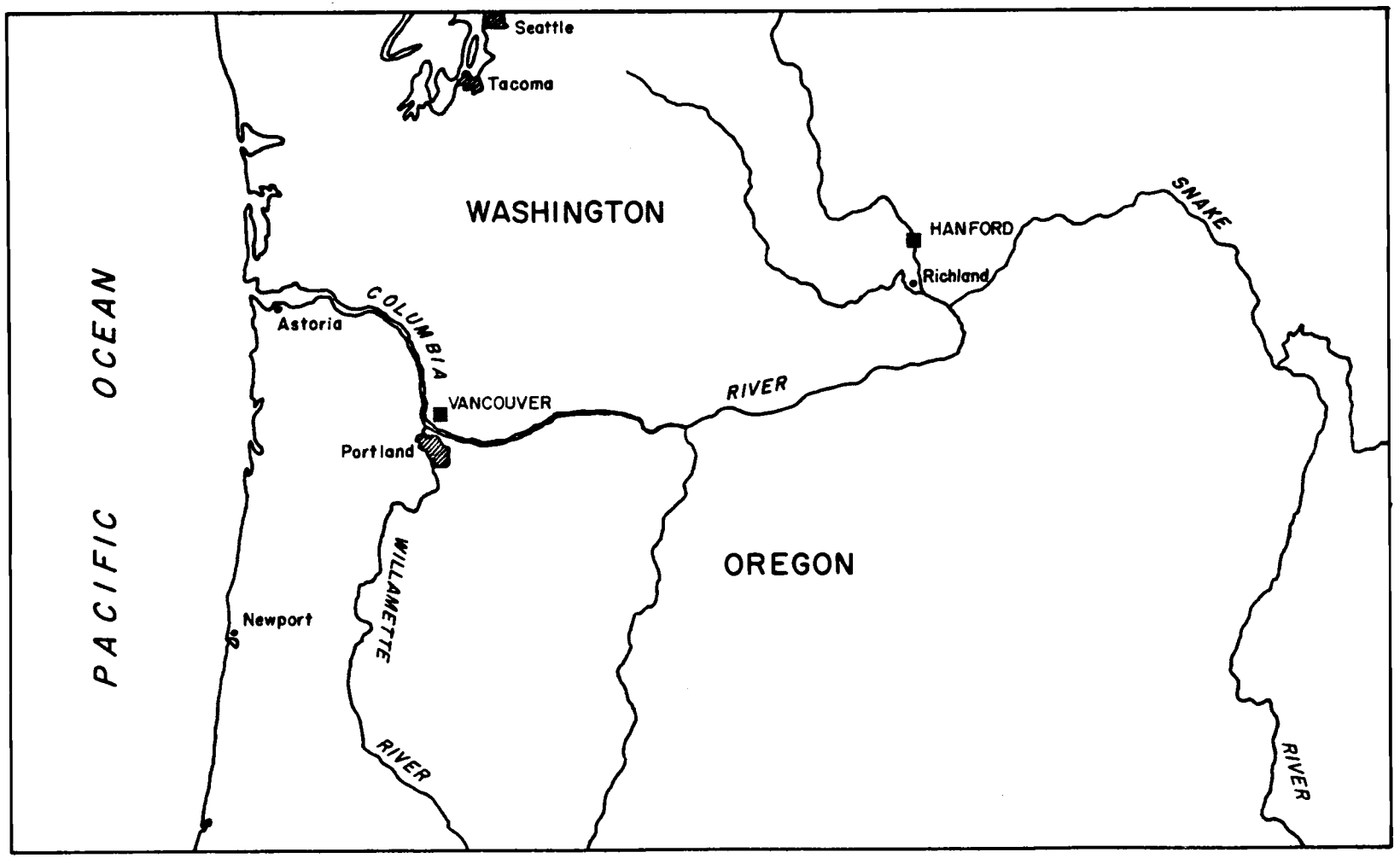

Figure 1. Columbia River Basin

It can be seen that P^{32} and Zn^{65} also are of sufficient quantity to be detected in the ocean, though they are not included in the list of relatively abundant radionuclides in the immediate vicinity of Hanford.

The rates of flow of radionuclides past Vancouver are actually of secondary interest. The most important values are the total amounts of radioactivity that actually reach the ocean and maintain themselves there as a result of the Hanford operations.* The radioactive material deposited in the ocean by the Columbia River is undergoing radioactive decay. If it is being added to the ocean at a constant rate over a period of time, eventually (depending on the decay constant and the rate of replenishment) equilibrium will be reached so that the radioactive decay of the nuclides in the ocean from Hanford just equals the rate of replenishment of these same nuclides from Hanford. These equilibrium values, computed for the year 1960 and based on the average flow past Vancouver, are (48, p. 22):

*The concentration per unit volume is perhaps of more interest, but this problem is exceedingly complex once the river empties into the ocean.

RADIONUCLIDE	APPROXIMATE EQUILIBRIUM VALUE, CURIES
P^{32}	360
Np^{239}	240*
Cr^{51}	33,000
Zn^{65}	14,000

*Using $N_o = N_e\lambda$ and solving for N_e, then $N_e = \frac{N_o}{\lambda}$. $\lambda = \frac{.693}{2.33} = .297$. Given that $N_o = 72$ curies a day, then $N_e = \frac{72}{.297} =$ $\sim$ 242 curies equilibrium value, which is the value given in the Hanford Report for the Pacific Ocean (48, p. 22). This really is an equilibrium value of Np^{239} given as if the Pacific Ocean were at Vancouver, or as if the rate of flow of Np^{239} past Astoria, Oregon, were the same as it is past Vancouver. However, Vancouver is some distance from the ocean (Figure 1). Assuming a transit time of 4 days, the equilibrium value of Np^{239} would diminish by a factor of $e^{-\lambda t}$ or $e^{-.297(4)}$, that is, to about 74 curies. The longer half-lives of the other three radionuclides make the equilibrium values given for them more valid.

The origin of these four radionuclides is due principally to the radiative capture of thermal or epithermal neutrons while passing through the reactors. The process is described in Appendix I.

B. Fission Products (Principally From Fallout)

The second major source of radionuclides in the Northeast Pacific Ocean is from atomic fission, usually occurring as the result of a nuclear detonation. Atomic fission refers to the breakup of a heavy nucleus into two or more medium-sized fragments. Division into two equal fragments is not very likely. Rather, asymmetric modes are favored, with the maximum fission yields peaking at atomic weights of 95 and 138, in the case of slow-neutron fission of U^{235}. The primary products of fission generally possess an excess of neutrons, and decay by successive β^- processes to stable isobars.* Chains with as many as six β decays have been established (29, p. 73). Regardless of the fissionable material used and the speed of the neutrons used to induce fission, a number of smaller elements are formed by fission. Most of these are highly unstable, often decaying too rapidly to be detected at any great distance from the blast. Others are produced in too low yield to be detected, unless the competition from the other radioactive elements can be removed. Competition is diminished by the passage of time, so that isotopes with long half-lives, which may have been

*Isobars are atomic species having the same mass number but different atomic numbers.

initially present in low yield, make up an increasing percentage of the total radioactivity at progressively later dates.

Three fission products which are of great concern to man are iodine-131, strontium-90, and cesium-137. None of these were encountered in the plankton, however. Iodine-131, though actively concentrated in the thyroid gland (28), has such a short half-life (8.0 days) that it is a threat only immediately after a nuclear detonation or accident. Also, it is volatile so the analytical processes used in this study would not be expected to show it, even if it were initially present in the plankton. Strontium-90, though it has a long half-life of 19.3 years, was not encountered because it is not a gamma emitter and is in solution in sea water (42, p. 18). Sr^{90} does not appear to have the same biological import in the sea as on land, for three reasons. Strontium is chemically similar to calcium, being only one period apart in the periodic table. Since the latter element occurs in the sea at a level of about 440 ppm. and is actually "preferred" to strontium by most marine organisms, little strontium is taken up. Prosser et al (56) have succeeded in depressing strontium uptake by increasing the calcium content.* Radiostrontium is of less concern in the sea

*Kornberg (40), however, points out that the passage of pairs of similar ions is complex, so that uptake of an ion may depend on the total amount of the ion, but may be fairly independent of the amount of a chemically similar ion.

because of isotope dilution (43, p. 118). Stable strontium is present in the sea at a level of 6 to 1300 times that of the naturally occurring forms of the other major radioelements represented in fallout. Finally, strontium is removed from the surface layers of the ocean by the purely physical processes of scavenging and coprecipitation.

Strontium-90 decays to yttrium-90, an extremely energetic beta emitter. Y^{90}, unlike Sr^{90}, is particulate in sea water, and may be concentrated by organisms feeding on particulate matter. Since Y^{90} is a new chemical species, however, conditions of chemical competition and isotope dilution are different for it, and its form in sea water may be of secondary importance. Its detection in this study was precluded because it is not a gamma emitter.

Cesium-137, like Sr^{90}, has a long half-life (33 years) and a high fission yield (its atomic weight is close to one of the peaks at 138), making it a potentially dangerous radionuclide. Some invertebrates seem to concentrate it (17), but it appears to be excreted rapidly by humans (61). Most cesium is in solution in sea water (42, p. 18), which is perhaps the most significant reason for the failure of plankton to concentrate it and thus allowing it to go undetected in this study. Cesium is chemically similar to potassium, though the relationship is not as close as with strontium and calcium, in that they are two periods apart in the periodic table. Rickard (61) states that Cs will

not substitute for K with fresh water algae. In the sea, however, stable potassium tends to depress the uptake of radiocesium (51), implying chemical competition. Cesium is taken up most efficiently by the foliage of desert plants, particularly in alkaline soils (61). Generally the evidence seems to indicate, according to Lowman (43, p. 128), that "in the case of Sr^{90} and Cs^{137} concentration occurs on land and exclusion occurs in the sea."

The principal fission products remaining 45 days after a nuclear detonation are the ones found in marine organisms off the Oregon coast. These are Zr^{95}-Nb^{95}, $Ru^{103-106}$, and Ce^{141}. Ce^{144}-Pr^{144} may be present but below the level of detection, since less than 2 percent of the nuclear adjustments result in gammas of sufficient energy to have been measured. The amount of naturally occurring zirconium-niobium, ruthenium, and cerium in sea water is low, and therefore isotope dilution would not play the same role as with strontium. The fission yield of these radionuclides is high, and they are all particulate in sea water. These factors make this group of fission products more available to marine organisms than other fission products.

Table I, taken from Brezhneva, et al (15), lists the relative activities (in percent) of some of the basic radioactive fission products. The relatively short half-lives of the elements most concentrated in the marine biosphere (Zr^{95}-Nb^{95}, $Ru^{103-106}$, Ce^{141}) give an indication

TABLE I

RADIOACTIVITY REMAINING AFTER A NUCLEAR DETONATION

Isotope	Gamma Energy Mev.	Time after Fission			
		45 d.	1 yr.	3 yrs.	10 yrs.
Sr^{90}-Y^{90}	None	0.51%	5.06%	19.4%	56.5%
Zr^{95}	0.23, 0.73	14.6	4.86	0.008	---
Nb^{95}	0.758	12.9	5.42	0.021	---
Ru^{103}-Rh^{103}	0.498	12.5	0.47	---	---
Ru^{106}-Rh^{106}	Many	1.04	5.81	5.87	0.16
Ce^{141}	0.146	10.6	0.11	---	---
Ce^{144}-Pr^{144}	Many (2.1;1.48; 0.69; 0.06) <2%	12.8	61.5	43.4	0.34
*Cs^{137}-Ba^{137m}	0.662	---	2.9	13.6	36.0
*I^{131}	0.364, etc.	5.6 (20 days)	---	---	---

* Taken from Chipman (17).

that the long range outlook for a non-radioactive food source in the sea is good. This is true partially because of the three dimensional aspect of the marine environment, which results in considerable dilution. Also, as will be indicated later, the discrimination against these fission products in the higher trophic levels insures that the levels of radioactivity from these sources will be quite low in the foodstuffs from the sea that have been generally consumed by man.

III CHOICE OF BIOLOGICAL MONITORS OF RADIOACTIVITY IN THE OCEAN

The results of radioanalyses of unsorted net plankton samples are difficult to interpret. Not only will the composition of the plankton vary with time and location, but often replicate tows contain different groups of organisms. An increase in radioactivity in the sample may be due to an actual increase in the amount of radioactive material in the water, or to certain plankton in the net which concentrate the particular radioactive isotopes to a greater extent than do other plankters. Possibly increased adsorption due to larger surface area has some effect also, although this problem is still not resolved. According to Carritt et al (16), microplankton seem to concentrate radioactivity to a slightly greater degree than do macroplankton, suggesting that physical adsorption is important. On the contrary, Bonham (11) stated that mesh

size did not influence the activity levels of his plankton samples.

To avoid these problems, various investigators undertook radioanalyses of single species of organism. In a study of the effects of the effluent of the Columbia River, oysters and mussels were used to trace zinc-65 (71), and it was shown that the radioactivity of these sessile organisms fell off rapidly with distance from the mouth of the river. Others continued this work, expanding the program to include algae, such as Fucus, Ulva, Porphyra, Gigartina, crabs, certain fish, bottom sediments, sea water and net plankton (63).

Only net plankton seems at all suitable for use as a biological monitor of radioactivity in the open sea. Most of the other organisms are either neritic or littoral. The fish are capable of movement in and out of areas of possibly high radioactivity, and may not reflect the radioactivity of their immediate environment. They also have the disadvantage of being at a high trophic level; that is, they would tend to discriminate against certain radioisotopes to a much greater extent than would plankton. Bottom sediments were excluded in this research a priori since they are primarily terrestrial in nature. Sea water, though considered elsewhere in this study, is not biological, of course.

While certain net plankton fulfill the requirement of being truly oceanic, the typical plankton tow yields an undefined assortment of organisms of various sizes and shapes and may contain representatives

of many Phyla. Largely because of this variety, the radioactivity of unsorted net plankton has been found to be 500 to 10,000 times that of an equal weight of surface sea water and is an extremely variable quantity (45). Clearly, unsorted net plankton would introduce too many uncertainties to make it of value as a biological monitor of radioactivity.

Rather large amounts of plankton are needed for radioanalysis, especially if only a small sorted segment of the complete haul is to be used. It was found that the half-meter nets and Clarke-Bumpus samplers customarily used in plankton programs were too small to provide the biomass required in single hauls. The advent of the routine midwater trawl program in the Department of Oceanography at Oregon State University furnished macroplankton in some quantity, however. Only macroplankton were considered in this study, as only these could be separated from the net plankton in sufficient quantity for analysis.

A choice of certain species of macroplankton from the trawl tows was required for radioactive monitoring, so that intercomparisons of samples from different areas of the ocean could be made. These intercomparisons would allow assessment of real differences in the amount of radioactivity in the marine environment. If identical species of organisms were not used, the same problems that arise with the use of unsorted net samples would occur.

A. Salps

A tentative choice of Salpa as a monitoring organism was made early in 1961. Salpa is a ubiquitous colonial tunicate whose abundance and size make it available in the quantities required. Furthermore, it was known to be primarily herbivorous and one of the great grazers of the sea (27, p. 1-2). This role as a filter feeder in the upper oceanic waters cast it as a potential concentrator of particles and ions from the many liters of sea water it is known to process daily. Particulate inorganic matter had been detected in its digestive tract (24), indicating the tendency to trap all particles in the water, whether edible or inedible. It appeared to be truly an indiscriminate feeder.

A sample of salps collected off Newport, Oregon, was taken to Hanford, Washington, for a preliminary analysis of radioactivity. The detection of zinc-65 and a suggestion of chromium-51 in this sample led to a search for additional salps to carry out the survey of radioactivity. The trawling program had already collected salps in quantity, and some of these were tested (see Results and Conclusions), verifying an earlier suggestion that zinc-65 was indeed ubiquitous in these waters (49), and the supposition that salps were good monitors of radioactivity.

Although salps were abundant in the summer of 1961 (see Appendix II), a good spatial distribution was not obtained. Most of the trawling operations were concentrated off Newport, as they were designed primarily for personnel training purposes. The need for the R/V ACONA to spend part of the early fall in drydock caused an interruption in the program, also. Because of the seasonal variation in the abundance of salps, this delay proved costly, since no more salps were observed in Oregon waters once operations were resumed until the cruise of March 8, 1962, when salps once again were collected. These factors, plus the early uncertainties concerning the role of the Hanford Laboratories in providing the instrumentation to make this research practicable, did not make it feasible to fully exploit the possibilities of using salps as collectors of radioactivity.

B. Euphausiids

The inability to find sufficient salps to provide the continuity required for monitoring purposes prompted consideration of a different organism, Euphausia pacifica. The change to a euphausiid for use as a monitor of radioactivity in the open ocean proved valuable for many reasons:

1. Our departmental observations show that E. pacifica is more nearly ubiquitous than Salpa, particularly during the fall, winter, and spring months.

2. E. pacifica is readily identifiable (8, p. 333-335) while the taxonomy of Salpa is confused (27, p. 1-4).

3. The euphausiid plays a more important role in the marine food web in that it is forage for many predators. Boden, et al (8, p. 288) state that "they are considered second in importance only to copepods as basic animal food in the sea but often exceed the copepods in mass and number, especially at greater depths."

4. Euphausiids are known to undertake vertical diurnal migrations (32, p. 199, 208). Our midwater trawl records show only sparse catches in the upper 200 meters in the daytime, while hauls made in darkness are often heavy. These migrations make the great euphausiid biomass a possible vehicle in the transport of radioactivity across the thermocline. Comparatively little transport would be expected by salps, which are weak swimmers. There is some indication that salps occur in somewhat larger numbers in the surface waters in darkness, but our evidence is limited.

5. While salps appear in two forms (solitary and in chains; i.e. asexual and sexual) according to Berrill (6, p. 151), the life

cycle of the euphausiid lacks this complication (7, p. 373-386).

6. Like the salp, E. pacifica is a second trophic level, particulate feeder (55).

7. E. pacifica has proven to be a better concentrator of radioactivity (on a dry weight basis).

The first euphausiids tested at Hanford were collected in November 1961. In addition to zinc-65, they contained several fission products attributed to airborne fallout from the Russian nuclear tests (50). Therefore, the bulk of the radioanalyses of plankton carried out subsequent to this first trial were of euphausiids.

C. Location of Radioactivity in the Organisms

Radioactivity, whether ionic or particulate, can be ingested or merely can be adsorbed to the outside of an organism. Since all radioanalyses were carried out on preserved plankton, their treatment before separation might alter the levels of radioactivity deposited on their outer surfaces. Martin (45) however, has reported that washing with uncontaminated sea water did not remove any detectable amount of the radioelements (including zinc-65, ruthenium-106, and zirconium-95) from foraminifera.

Zinc-65, the most abundant radioisotope encountered in plankton before Russian fallout, is a transition element. According to Lowman (43, p. 117), transition elements become complexed to exposed polar groups found in organic detritus and microorganisms. These are the groups which also form complexes with anions such as zirconium and ruthenium, etc. It is extremely difficult to remove a complexed ion by washing; for example, Goldberg (30, p. 352) states that much mineral matter can be washed away without a loss of trace elements. A priori there seemed to be no reason to expect a loss of zinc from the organisms in this study. The evidence for particulate fission products, such as Zr^{95}-Nb^{95} and Ru^{103}, seemed less convincing, however, and adsorption might be important where surface areas are large. Compared with large numbers of small organisms in unsorted net plankton, the surface areas of euphausiids (about 1 inch long) and salps (1/2 to 6 inches) are relatively small per unit of mass.

The effect of adsorbed radioactivity could be resolved partially by making radioanalyses of the inner and outer parts of the macroplankton separately. This was clearly possible with salps but not with euphausiids. Salpa has a round dark "nucleus" which contains the digestive tract. Most of the muscles of propulsion and remaining jelly-like material making up the transparent protoplasm can be removed easily from the "nucleus." It is the transparent protoplasm

which is in contact with sea water and hence subject to adsorption of radioisotopes. The salps chosen ranged in size from about 1 to 3 inches. The "nuclei" made up 6.5 percent of the wet weight of the salps and 13.3 percent of the dry weight; the balance of the weight was in the outer transparent portion. The results of these radioanalyses are shown in Figure 2.

The concentration of radioactivity in the "nuclei" is much greater on a weight basis than it is in the exterior protoplasm. Furthermore, the total radioactivity in the "nuclei" is greater than in the much heavier transparent portion. Thus, it appears that surface adsorption of radioisotopes plays only a very minor role, if any, in the concentration of these radioactive materials by salps.

A comparison of the ratios of picocuries per gram dry weight of "nucleus" to picocuries per gram dry weight of exterior protoplasm is shown below:

ISOTOPE	SALP NUCLEI	SALP EXTERIOR	RATIO OF NUCLEI TO EXTERIOR
K^{40}	56.0	6.6	8.5 : 1
Zn^{65}	18.9	1.6	11.8 : 1
Ru^{103}	56.2	1.8	31.2 : 1
Cr^{51}	172.2	5.5	31.4 : 1
Zr^{95}-Nb^{95}	288.7	4.1	70.5 : 1
Ce^{141}	126.8	0	∞

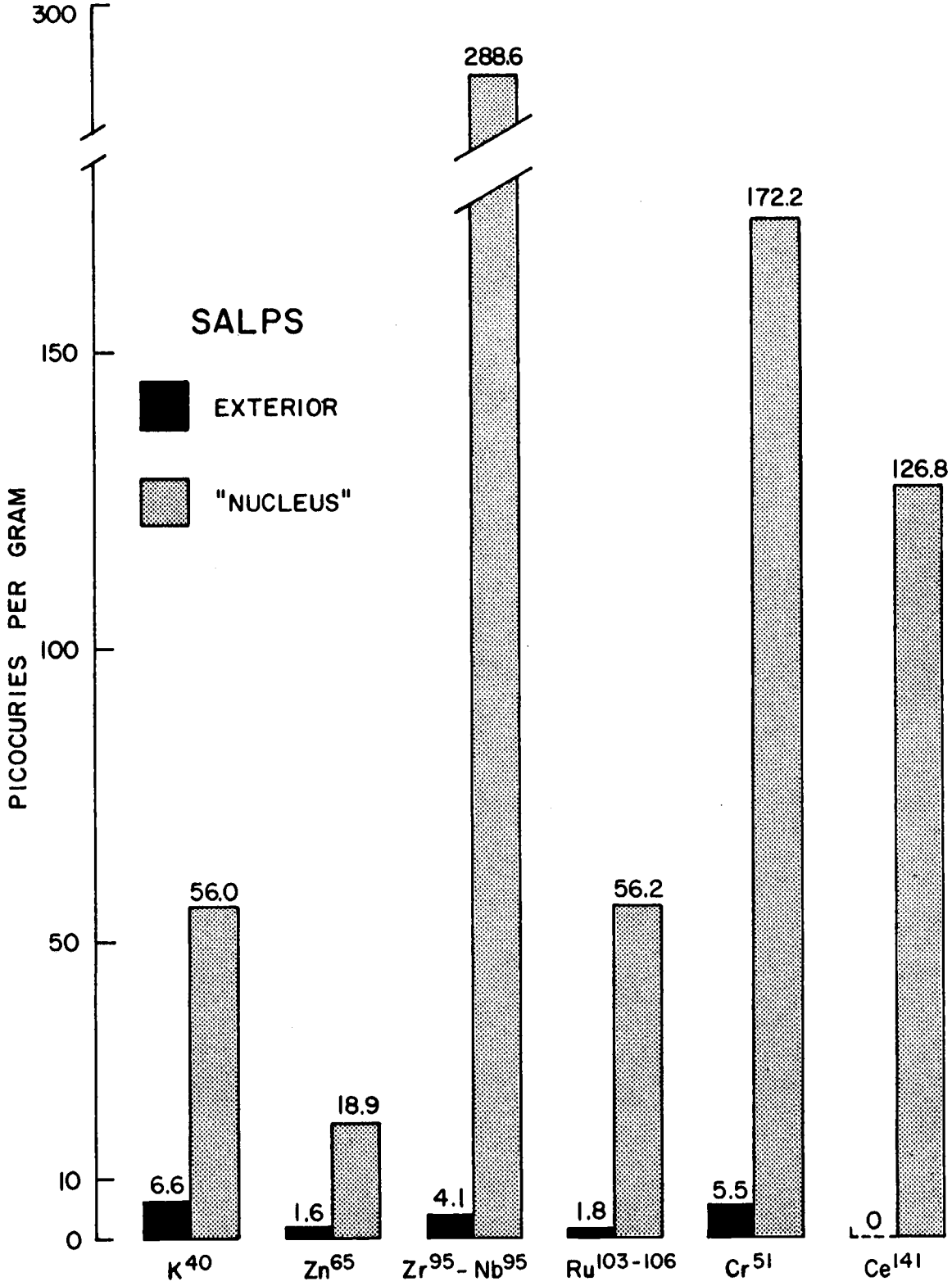

Figure 2. Radioactivity of salp interior compared with exterior

The ratios appear to fall into groups. K^{40} and Zn^{65}, presumably non-particulate (and cations) in sea water, are least concentrated in the gut (nucleus). Zr^{95}-Nb^{95} and Ce^{141}, probably particulate (and anions) in sea water, are most strongly concentrated in the gut. Ru^{103} and Cr^{51}, whose physical form in sea water is less well known (anions), appear intermediate. The extremely low radioactivity of the exterior protoplasm makes the ratios subject to large variations, however.

It should be mentioned that the much greater radioactivity of the "nuclei" does not necessarily imply assimilation, since very possibly the radioisotopes are confined principally to the intestinal tract, and thus would not be an integral part of the organism. This would be of no consolation to a predator feeding on the salp since in any case the radioactivity, whether externally adsorbed or internally assimilated by the salp, would be "taken internally" by the predator.

A second experiment was conducted to provide further information on whether the radioactivity was due to adsorption and thus a function of surface area. Copepods are closely related to euphausiids (they are both pelagic crustaceans) but are smaller, and therefore, have a much greater surface area to weight ratio than do euphausiids. A single sample was chosen which contained both

E. pacifica and copepods in sufficient quantities for radioanalysis. The copepods were about 95 percent Calanus cristatus (by volume), with the balance almost exclusively Calanus plumchrus. The results of this comparison are shown in Figure 3. Although the sample was collected 105 miles off Astoria in a region of low radioactivity, yielding relatively poor statistics, there is little doubt that the tremendously greater surface area of the copepods has had no enhancing effect on their radioactivity. Rather, it appears that euphausiids, per gram dry weight, are somewhat better concentrators of these particular radioisotopes. Again, the conclusion is that surface adsorption has been of no apparent importance in the uptake of radioactivity by these macroplankters.

IV. MATERIALS AND METHODS

A. Sample Collection and Preparation

1. Macroplankton and Nekton

Most of the organisms were obtained by routine trawling operations off the Oregon coast with a modified 6-foot Isaacs-Kidd midwater trawl (34, p. 1-18). The stations regularly visited are

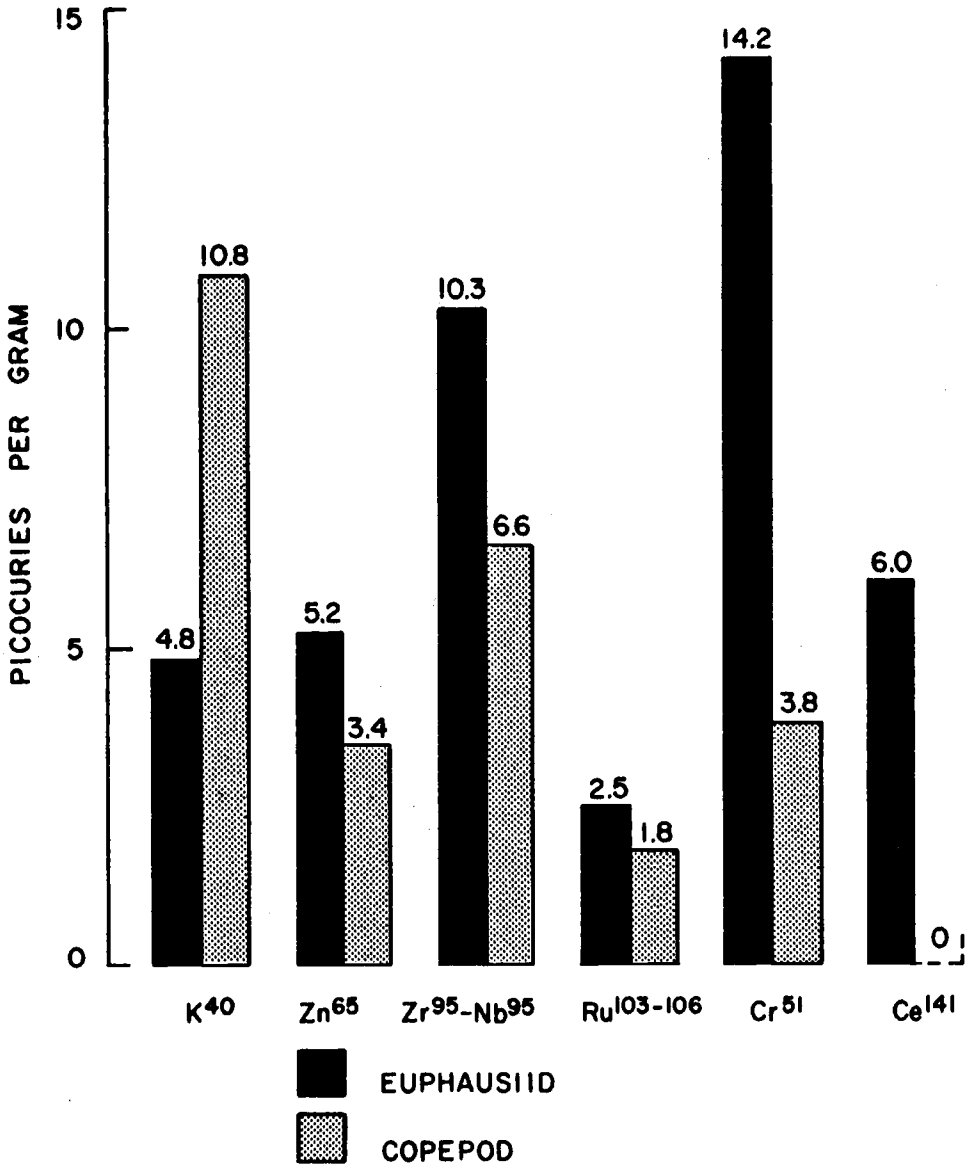

Figure 3. Comparison of radioactivity between euphausiids and copepods from the same tow

shown in Figure 4. Fishing time at depth was normally 30 minutes, and trawl depth was 200 meters (depth permitting).

Samples collected in the trawls included a mélange of organisms ranging from large copepods to small fish and squid, with euphausiids and sometimes, salps, making up the bulk of the biomass. All samples were preserved in buffered formaldehyde aboard ship. After various groups of larger organisms were removed for nekton studies, the balance of the collection was inventoried and stored. Salps, and to a much greater extent euphausiids, were separated from this supply of preserved plankton in the laboratory for radioactivity monitoring purposes. Separation often was tedious, especially when collection contained an abundance of gelatinous organisms, or when the quantity of the desired macroplankter was barely sufficient for radioanalysis. Some tedium also was experienced in samples which contained Euphausiacea other than E. pacifica because of the time needed for visual inspection. In general, however, E. pacifica was the dominant euphausiid, and sometimes appeared in such abundance that almost no sorting was required. No effort was made to separate the salps by species, and sexual and asexual forms were combined in the sample. Identification of subsamples subsequently showed that

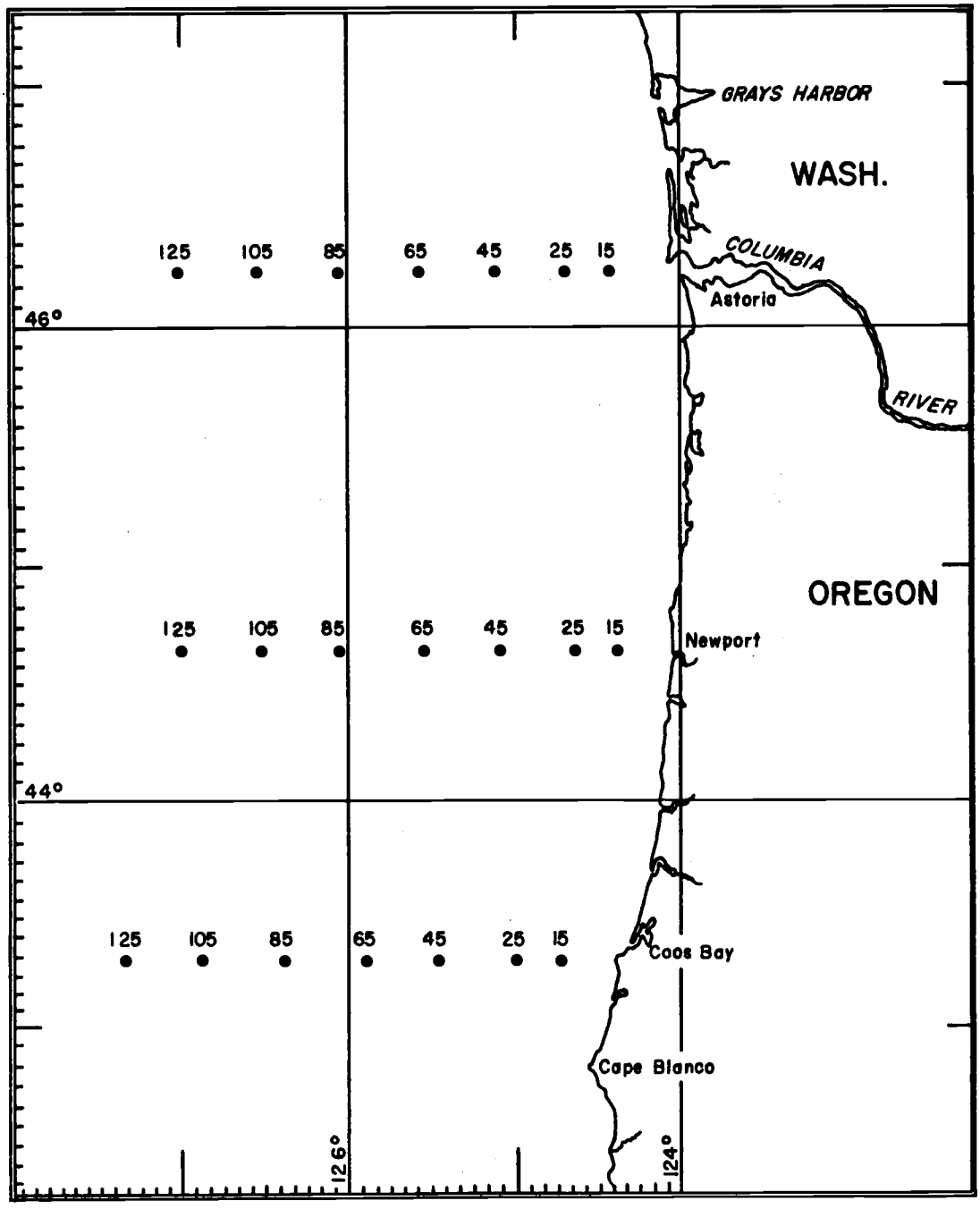

Figure 4. Map of midwater trawl stations

the salps were principally Salpa fusiformis and Salpa aspera. Iasis (Salpa) zonaria was found frequently but never in quantity. Certain other organisms taken in the nets were preserved for radioanalysis, also. Some were predators which feed on euphausiids and thus were a part of the same food chain. In this category were the lantern fish, Lampanyctus leucopsarus, and the viper fish, Tactostoma macropus. The latter, in particular, may also feed on small predators so that the food web relationship is uncertain. The sergestid shrimp, Sergestes similis, and the carid shrimp, Pasiphaea spp., also were analyzed for radioactivity, mostly on a trial basis. A subsample of Pasiphaea showed that P. pacifica was more common than P. magna, but the single sample which was analyzed is not conclusive evidence of general abundance.

While sample collection and separation of macroplankton often were difficult and time consuming, the method of preparation for radioactive counting which proved suitable for most of the samples was fast and simple. Since most marine organisms contain large amounts of sea water, it is desirable to remove this before radioanalysis. The water is not radioactive (at least no isotopes of hydrogen or oxygen are gamma emitters), so that the removal of this large volume of non-radioactive material by drying

concentrates the radioactivity in the residual dry portion. Drying was carried out by the freeze-dry method. The samples were removed from the formaldehyde solution, drained briefly, then weighed. They were then placed into shallow 8 x 12-inch Pyrex glass trays which had been coated previously with Beckman New Desicote to prevent sticking. The samples were frozen at - 18°C., then dried in a vacuum oven, which could accommodate 10 trays at a time, in which the heating coils were maintained at about 130°F. The air pressure was reduced to .4 to .7 mm of mercury and, even with the heating coils, the samples remained frozen at this pressure as long as they contained water. Drying usually was accomplished overnight. If further concentration of the samples than could be obtained by the freeze-drying technique was necessary, or if sample size permitted, the samples were ashed in a muffle furnace. Both techniques were used in this study.

After drying, the samples were ground with a mortar and pestle and placed into 17 x 100 mm pre-weighed counting tubes.* Because of the texture of the dried, pulverized plankton, it was packed into the tubes a little at a time, using a rounded wooden

*Falcon Plastics, Item number 2001.

rod. This increased the specific activity per unit volume. Sample volume for the special Hanford counter, which will be described later, was 13 cc. While samples of other sizes could be "counted," the standardization and calibrations were based on a 13 cc sample. A lesser (or greater) sample would have different geometry and the accuracy of the count would be affected in an unknown manner. Samples analyzed in this study conformed to the 13 cc volume restriction.

It should be noted here that the freeze-drying and grinding procedure did not work well with fish. Lampanyctus leucopsarus, in particular, was rich in oil. After freeze-drying, its weight was still about 33 percent of its wet weight. The oil prevented grinding with mortar and pestle, and further treatment in the muffle furnace was required. After ashing, the weight dropped to about 5 percent of the wet weight. No effort was made to ash to "constant weight;" rather, proper geometry was striven for by filling the counting tube to the prescribed 13 cc mark. This was accomplished rather easily because of the natural compressibility of the material. The minor changes in density were considered not to influence the accuracy of the counting because of the small sample size and penetrating ability of the gamma photons

concerned.

After filling to the 13 cc volume, all sample tubes were reweighed, and the net weights of the samples were obtained. Each tube was sealed with an inverted #4 cork so that the large end of the cork rested against the compressed sample. Melted paraffin wax was poured around the tapered end of the cork, filling the tube completely. After the wax had hardened, the plastic lid was forced on tightly. This process sealed the tube against moisture, possible spillage, and changes in geometry. The sample was then ready for radioactive counting.

2. Particulate Matter and Phytoplankton

In addition to the trawl samples of macroplankton, an effort was made in the spring of 1962 to sample the particulate matter in the ocean for radioanalysis. Since euphausiids and salps are particle feeders, a membrane filter would be expected to duplicate the ability of these macroplankton to remove particulate material from the water. Particulate material would of course include many of the fission products, as well as phytoplankton. Radionuclides that are ionic in sea water would, on the other hand, pass through the filter unless they were associated with particulate matter. To

give a rough indication of the percentage of phytoplankton contained in the total particulate matter, an analysis for chlorophyll a was made. A subsequent comparison of the radioactivity of the filter with that of the macroplankton gave some idea of the ability of the macroplankter to concentrate, or to discriminate against, a radioisotope.

The filtering program was carried out by using a suction pump to pull sea water, collected from the surface, through a pre-filter and through a 5-inch membrane filter designed to trap 90 percent of all particles larger than .1 micron. The pre-filter (a 5-inch glass fiber filter) was used to prevent too rapid clogging of the membrane filter and give "filtration in depth."* With this setup, a minimum of approximately 35 liters of surface sea water was filtered at each of several stations. Zooplankton were removed before filtration by straining the water through #6 mesh (aperture size, 0.239 mm). The friability and consequent loss of edges of the glass-fiber prefilter precluded the determination of radioactivity on a weight basis, so that results had to be expressed per unit volume of water.

*Both filter and prefilter were obtained from the Gelman Instrument Co., as was the filter holder.

The filters did not require freeze-drying as the macroplankton samples did, but rather were dried in a dessicator. Efforts to grind the membrane filter and glass-fiber prefilter were not successful. It was found that about 30 minutes of heating in the muffle furnace at 300°C would destroy most of the membrane filter and soften the glass-fiber filter so that they then became easily friable in the mortar. The complete procedure developed for the filters involved cutting the membrane and glass prefilter (along with the trapped particulate matter) into small sections and placing these sections directly into a crucible. A lid was placed on the crucible and it was put into the muffle furnace. After incineration and cooling, the contents of the crucible were ground in a mortar with 10 cc of sucrose which is low in radioactivity and has a density comparable to plankton.* The sugar seemed to aid the grinding process and also served to give the sample the correct volume to conform to the prescribed counting geometry. Mixing while grinding was sufficient to assure a homogenous sample. After grinding, each sample was packed

*When an attempt was made to process several filters in the same crucible, the mixture erupted mildly when the kindling temperature was reached, scattering the contents. Therefore, subsequent samples were burned with an open flame before being placed in the muffle furnace.

into a 13 cc counting tube, and the tubes were sealed exactly as the macroplankton sample tubes.

B. Radioisotope Counting Techniques

Scintillation spectrometry, the method employed in this study to evaluate gamma radiation from the various samples, has been more of an art than a science. No two crystal-photomultiplier-spectrometer combinations appear to have the same response to a source of radioactivity. Despite the expense and capriciousness of this tool, however, the number of gamma-ray spectrometers in use has grown rapidly.

The instrument used in this study is probably the best in the country for working with low levels of radiation when only a very small sample is available. The system, called the Hanford Total Absorption Anticoincidence Spectrometer, was made available by its principal designer, R. W. Perkins, at Hanford Atomic Laboratories, Richland, Washington. It consists of a 5-inch diameter by 5-inch thick NaI(Tl) crystal with a 7/8-inch diameter by 3-inch deep well. This crystal is optically coupled to four 6655A photomultiplier tubes. A second phosphor, to detect degraded photons from Compton scattering or annihilation

radiation escaping from the main crystal, also is employed. It is a plastic 26-inch diameter, 30-inch thick cylinder (in three pieces) with a 6-inch diameter, 12-inch high chamber inside it to accommodate the primary crystal. The large secondary phosphor is viewed by twelve 5-inch DuMont 6364 photomultiplier tubes. The complete detector is surrounded by a 4-inch thick lead shield with an opening in the top to permit placing the 15 cc plastic counting tube (with a 13 cc sample) into the well of the primary crystal. The signal from the detector is fed through an amplifier to a 400 channel RIDL pulse height analyzer (53).

It is difficult to compare results obtained with this instrument with those obtained with less sophisticated detectors, since radioanalyses have not been carried out by other instruments on any of the organisms investigated herein. The Hanford instrument is vastly superior to those using a single crystal in counting such radionuclides as Ru^{103} (0.498 Mev gamma) in the presence of the overlapping annihilation peak of Zn^{65} (0.511 Mev). Figure 5a shows that the anticoincidence feature of the Hanford instrument reduces the annihilation peak to a point where almost no interference results in the region of the Ru^{103} peak

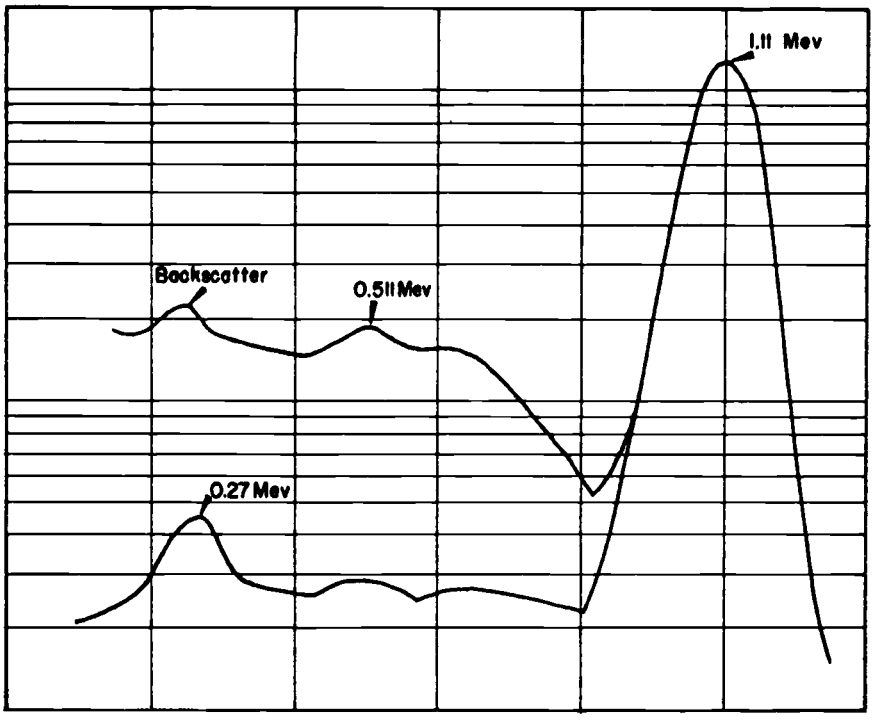

Figure 5a. Zinc-65 spectrum showing annihilation peak

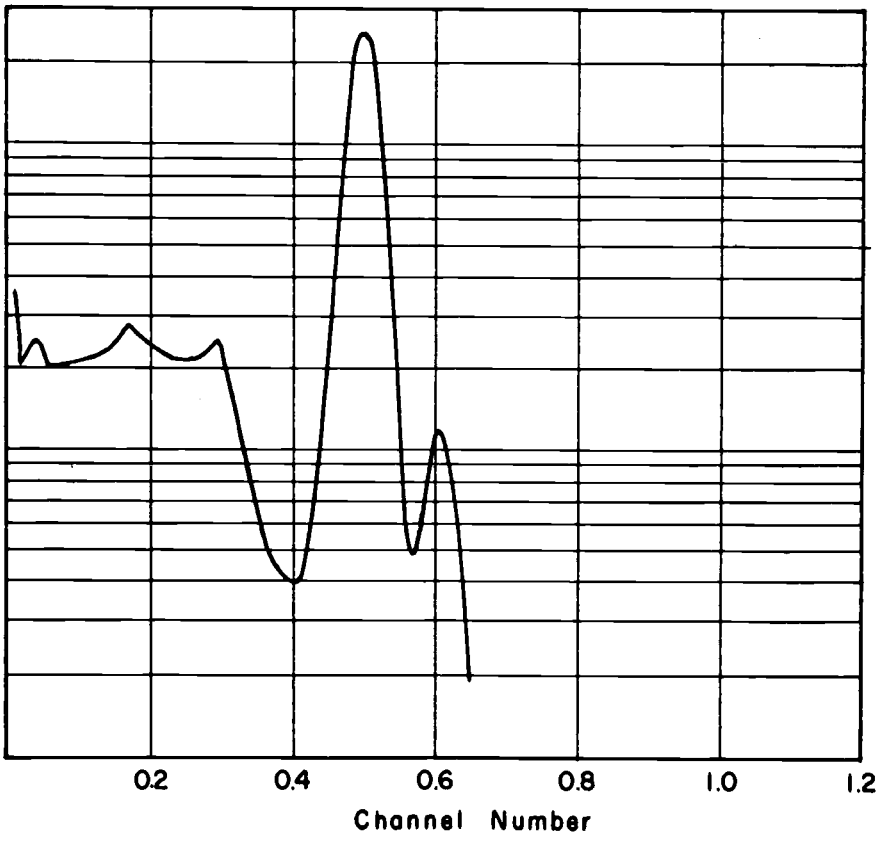

Figure 5b. Spectrum of ruthenium-103

(Figure 5b). This reduction in background gives a higher signal to noise ratio, which results in greater accuracy.

The plastic anticoincidence secondary crystal has several objectionable features, namely, its high counting rate of background noise, its expense, its bulk (requiring a large amount of shielding), and most important, its poor resolution. Nevertheless, it is felt that in overall consideration no other detector is as well suited for research on radiation in small quantities of marine organisms as the one used in this study.

The RIDL multichannel analyzer used with the described detector, like most electronic equipment, tended to drift. This drift can be seen in a comparison of two trial gamma ray spectra of euphausiids collected 15 miles off Astoria, Oregon, and 15 miles off Coos Bay, Oregon (Figure 6). The 50-minute counting time clearly resolved the Zn^{65} and the Ru^{103} bands in both spectra and showed an indication of some Cr^{51} at Astoria and Zr^{95}-Nb^{95} at Coos Bay, but drift is particularly evident in the Ru^{103} band, the peak of which is displaced about one energy channel upward in the count of the Coos Bay sample (this drift is purposely exaggerated in the drawing). Because of this drift, all subsequent counts were made with the analyzer adjusted

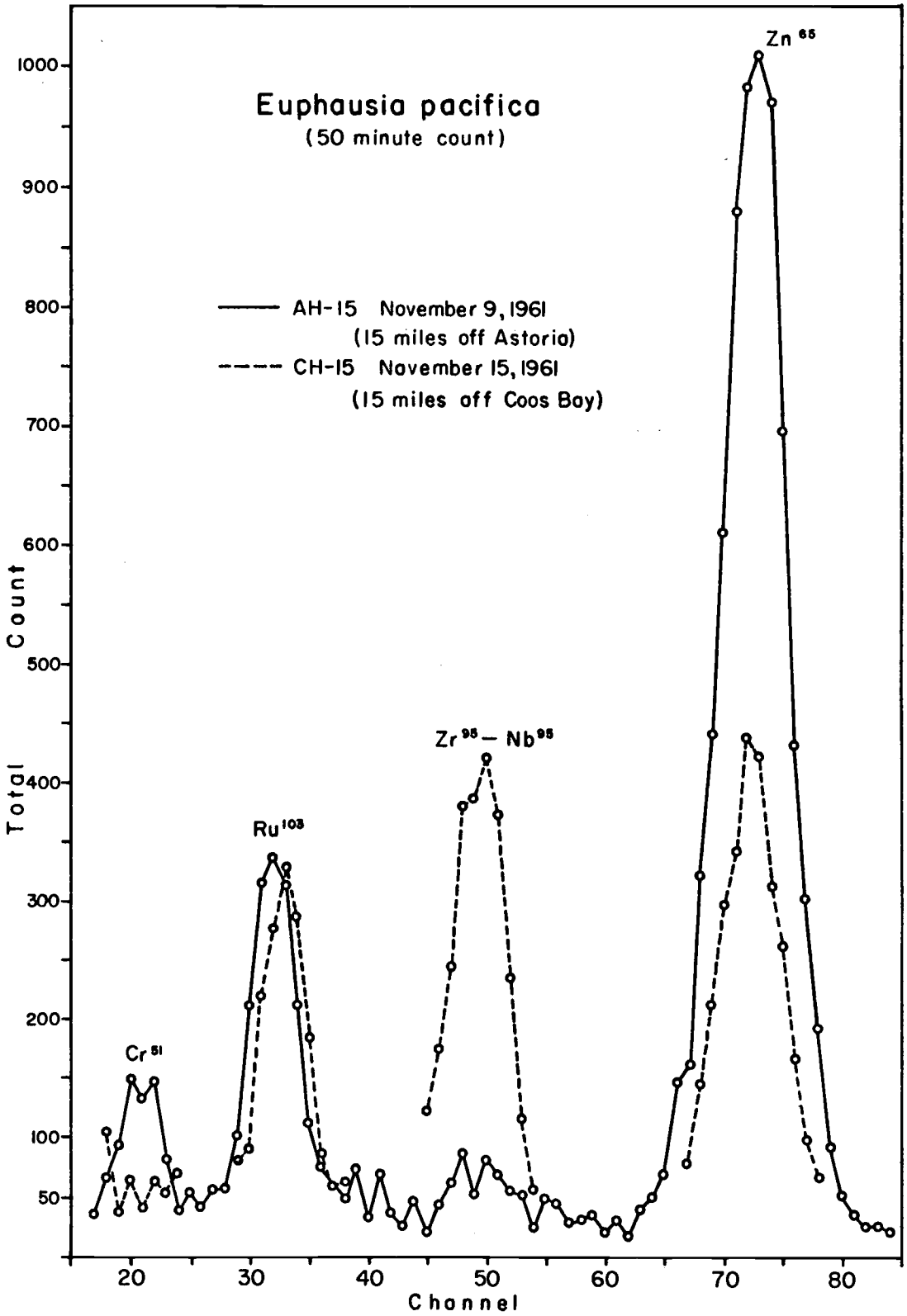

Figure 6. Comparison of spectra from different locations

before each counting period, so that the 0.662 Mev photopeak of a calibration source of Cs^{137} would center in channel 44. Zero was set between channels 10 and 11 and between channels 100 and 101. This procedure effectively compensated for the channel drift.

C. Data Reduction

To obtain meaningful data from the spectra obtained from a typical mixture of radionuclides in plankton taken off the Oregon coast, it was necessary to separate the effects of the spectrum of each element from all other spectra. Only then was quantitative analysis possible. To accomplish this, knowledge of the response of the spectrometer used in this study to gamma emitters of known activity was required. Calibration of the spectrometer was accomplished by the personnel at Hanford and will not be discussed in detail here since the information is readily available elsewhere (52; 19).

Table 2 summarizes the numerical constants unique to this instrument, which were obtained in the calibration, however. They are used to convert counts integrated through the area of the photopeak (i.e., the sum of the counts in the individual channels which comprise the photopeak) into picocuries (micromicrocuries). The column "Channels Used" defines the limits of the photopeak for the isotope under examination. For example, potassium-40 has a wide

TABLE 2

INSTRUMENTAL CONSTANTS USED WITH HANFORD TOTAL ABSORPTION ANTICOINCIDENCE SPECTROMETER

Isotope	Channels Used	Efficiency Factor	K^{40}	Zn^{65}	Zr^{95}-Nb^{95}	Ru^{103}-Rh^{103}	Cr^{51}
K^{40}	286-303 (1.46 Mev)	40.55					
Zn^{65}	268-279 (1.114)	6.00	0.0440				
Zr^{95}-Nb^{95}	246-255 (.72,.75,.77)	2.480	0.0375	0.0459			
Ru^{103}-Rh^{103}	230-238 (.498,.610?)	4.037	0.0553	0.0480	0.0718		
Cr^{51}	219-225 (.32)	14.71	0.113	0.0528	0.0682	0.0898	
Ce^{141}	209-211 (.145)	3.054	0.133	0.0117	0.0192	0.0417	0.0358

5 inch well with coincidence; Cs^{137} in channel 44; zero 10-11, 100-101; 13 ml in plastic tube

peak extending from channels 286 through 303. Cerium-141 has a very narrow peak extending only from channel 209 through 211. In part, these peak characteristics come about through statistical considerations of the gamma energies of the isotopes. Each channel of the analyzer is about 0.015 Mev wide. The energy of the K^{40} peak is 1.46 Mev while the peak for Ce^{141} falls at 0.145 Mev. Thus, more 0.015 Mev channels are required to show the K^{40} peak than are required to describe the Ce^{141} peak.

The third column of Table 2 is the efficiency factor for each radionuclide. This factor is determined by the energy of the gamma photon of the radionuclide in question, the decay scheme of the nucleus, geometry, and crystal size. For example, the efficiency of counting high-energy gammas is lower than that of low-energy gammas because more high- than low-energy gamma photons escape the crystal. Also, a correction must be made if for example, in the decay scheme of a nucleus only one disintegration in three (on the average) leads to a gamma photon. Geometry refers to the portion of gamma photons which actually strike the crystal. For a point source, this is the fraction of the total solid angle, 4π, which the crystal subtends at the source. A sample placed on the surface of a crystal (as in simple detector-analyzer arrangements) yields approximately 2π geometry; if it is placed into a well, the geometry theoretically can be increased to

4π. In practice, these limits are not attained; therefore, the geometry of the sample should duplicate that of a standard. If it does not, the efficiency factors will not be valid. The efficiency factors in Table 2 were based on a sample occupying 13 cc in a 17 x 100 mm plastic tube. Care was taken in this study to meet this requirement.

The efficiency factor is a function of the ratio of absolute disintegration rate to count rate recorded in the spectrometer. K^{40}, for example, has a high-energy (1.46 Mev) photon with great penetrating power, and since only about 11 percent of the disintegrations yield this countable photon (39, p. 217) and many of these escape the crystal, only one nuclear disintegration in 40.55 is recorded; therefore, the final number of counts in the photopeak must be multiplied by this factor. Zn^{65}, with 49 percent of the disintegrations yielding a 1.114 Mev gamma photo (66, p.27), must be multiplied by 6.0. The same reasoning is valid for Cr^{51} and Ce^{141}.

Zirconium-95 and niobium-95 present a special problem; 45 days after a nuclear detonation, they make up 14.6 and 12.9 percent, respectively, of the total radioactivity remaining as a result of the blast (15, p. 220). Because of the longer half-life of Zr^{95} (65 days compared with 35 days for Nb^{95}), the activity of Nb^{95} might be expected to decrease more rapidly. However, Zr^{95} decays

by beta emission to become Nb^{95}, so that actually the activity due to Nb^{95} with respect to Zr^{95} increases with time. After one year, the activity due to Zr^{95} and Nb^{95} is 4.86 and 5.42 percent, respectively (15, p.220). Therefore, the two isotopes must be evaluated together as Zr^{95}-Nb^{95}, and an efficiency factor of 2.480 is used for the unknown mixture of the two which is found in nature.* The difference in half-lives of the two isotopes is too small for this relationship to be considered secular equilibrium (see Rock et al (62) for complete discussion of this decay curve). The best approximation that can be given is that the mixture of these two fission products is in equilibrium in the plankton and decays at the rate determined by the longer half-lived parent, i.e. 65 days. It is apparent that some errors would be introduced if it should turn out that the organisms selectively take up niobium and exclude zirconium; however, there is no good physiological evidence that this is true. On the contrary, Zr^{95} appears to be preferred by invertebrates (45). Only a chemical separation before gamma spectrometry can resolve the problem, since

*Generally Nb^{95} is used in the calibration since there is no question as to its purity and therefore its half-life. Its energy peak and decay scheme is so similar to that of Zr^{95} that this substitution is possible. If pure Zr^{95} were used, an increase in activity (due to formation of Nb^{95}) would be experienced with the maximum activity occurring 25 days later (62).

the spectra of Zr^{95} and Nb^{95} are almost identical (20, p.200).

The remainder of the columns of Table 2, labeled K^{40}, Zn^{65}, etc. define the percentage of the area of the photopeak which is due to the Compton continuum from another radionuclide of higher energy. Thus we see that every 100 counts in the K^{40} photopeak will give 4.4 counts in the area of the Zn^{65} peak. This amount must be subtracted from the total number of apparent counts in the Zn^{65} band. Figures 5a and 5b show that the Compton continuum from Zn^{65} falls in the area of the photopeak of Ru^{103}; therefore, its effect must be subtracted from the area of the Ru^{103} photopeak before quantitative results are possible. It should be noted that the reverse effect is negligible, since the Zn^{65} photopeak falls in a region essentially free from the influence of Ru^{103}. A correction for Ru^{103} therefore is not needed in the calculation for Zn^{65}. The same is true regarding other radionuclides whose photopeaks fall at a lower energy than does Zn^{65}. The Ce^{141} photopeak, lying at the lower end of the spectrum, contains Compton scatter from K^{40}, Zn^{65}, Zr^{95}-Nb^{95}, Ru^{103}, and Cr^{51}. Because of this, spectrographic analyses for Ce^{141} and other low-energy gamma emitters are quite involved.*

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*Actually, the corrections are less for Ce^{141} than for Cr^{51}, as evidenced by Table 2. The explanation for this is beyond the scope of this paper, but the result is that the errors in measuring chromium-51 are more severe.

Table 3 gives an example of the computations required to arrive at the quantitative amounts of the six radioisotopes. Most of the early data were reduced using this computing form. When it became apparent that the radioanalysis program was to be continued and enlarged, a more error-free and less tedious method of data reduction was devised using an IBM computer available on the campus. The results, presented in columnar form, on the typewriter "read out", include the name of each of the six radioisotopes, the number of picocuries per unit of weight (or volume), and the standard deviation of the result. The raw data is also printed along with the results to facilitate checking for possible errors.

Considerable caution must be exercised in data reduction, since the results are invalid if isotopes other than the six programmed for are present. The errors become increasingly serious the higher the energy of the unknown contaminent, and are particularly large in the low-energy region of the spectrum. In other words, very few contaminents would effect the region of the Zn^{65} peak (1.114 Mev), whereas the majority of them would cause Compton scatter in the region of Cr^{51} (0.32 Mev). Because of the contamination possibility, it is preferable to have a trained observer inspect the gamma spectrum before the areas of the peaks are computed to make sure extraneous peaks are not present.

TABLE 3

COMPUTING FORM USED IN DATA REDUCTION

SAMPLE _____

LOCATION _____ DATE COLLECTED _____

5" well with coincidence, Cs^{137} in Channel 44, 13ml sample

Isotope	Aliquot	Efficiency factor	Observed c/m	Compton corrected	Date Time	Elapsed time	Decay Correction	d/m Corrected d/m	d/m/g	Picocuries/g
K^{40}		40.55								
Zn^{65}		6.00								
Zr^{95}		2.48								
Ru^{103}		4.037								
Cr^{51}		14.71								
Ce^{141}		3.054								

The statistical analysis as programmed on the IBM 1620 included the errors imposed on the area of the photopeak by the uncertainties in the Compton continuum from gamma emitters of higher energy. It should be emphasized also that the standard deviation obtained from the statistical analysis only pertains to the counting statistics. All other errors in sampling, evaluating efficiency constants, etc., are not considered and may be much greater. An example of the computations carried out in spectrum analysis and in determining the standard deviation is shown in Appendix III.

RESULTS AND CONCLUSIONS

A. Tracing the Columbia River by Zinc-65

It has been known for some time (4, p. 588) that the outflow of the Columbia River dilutes the salinity of the surface water in certain sections of the ocean, creating a shallow lens of less saline water which overrides the more dense oceanic water. In the summer, the prevailing winds carry this less saline water southward, sometimes to form a huge pool of about 100,000 square miles in area off the Oregon coast (2). The stability of the surface layers of the ocean in summer caused by increased insolation, calmer meteorological conditions and higher runoff of the river, favors this pooling. Identification of the

Columbia River "plume" as delineated by a salinity of 32.5% or lower (3, p. 6) is quite feasible at this time. In winter, however, decreased runoff and increased mixing by storms make identification of the river water at sea by the salinity method more difficult.

It was pointed out that an equilibrium value of about 14,000 curies of Zn^{65} is maintained somewhere in the Pacific Ocean as a result of the nuclear reactors at Hanford Laboratories. This constant outflow of Zn^{65} makes it possible to use the Columbia River in a huge radioactive tracer experiment. In view of the sensitivity of the radioactive tracer technique, Seymour (64; 70, p. 10) made an effort to correlate the distribution of Columbia River water at sea with the Zn^{65} contained in net plankton. Although some success was claimed for tracing the river by this means, no further reference beyond the mentioned unpublished report appeared in the literature. Plankton undoubtedly will concentrate the radioactivity and make detection of the Zn^{65} easier, but for reasons already mentioned, mixed plankton samples generally would be poorly suited to the task of monitoring the river. Radio-zinc in the water itself would be extremely difficult to detect any distance from the mouth of the river, because of the great amount of dilution by the sea water. The use of salps and euphausiids was regarded as a potentially more desirable monitor of the Columbia River water than either mixed plankton or the river water itself.

Even with the use of a single macroplankton species as a tracer organism, there existed the possibility that not all of the zinc-65 detected in the organisms would be attributable to the Columbia River. It is known that Zn^{65} was produced by the Pacific nuclear tests in 1954 (43, p. 126), and has been found in organic material even in the midwest (47). Although it is not listed among the possible radioisotopes produced by the N. S. Savannah (57, p. 24-25) or the U.S.S. Nautilus (57, p 26), oddly enough it has been found in some quantity in the oysters from the Thames River (Conn.) in the vicinity of an atomic submarine base (25). Zn^{65} is not a fission product*, but is produced by neutron activation (see Appendix I); thus it is possible that a nuclear device, whose structural parts contained much zinc, could contribute an appreciable quantity of Zn^{65} to the fallout. Because of the series of nuclear tests engaged in by the Russians beginning in September 1961 and the increasing use of nuclear submarines, and particularly because of strong peaks of fallout fission products often present in the spectra recorded in this study, there is the possibility that not all of the Zn^{65} in the Northeast Pacific originates at Hanford.

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*The minimum atomic number (Z) obtainable in fission is 30 (zinc), but the minimum atomic weight (A) is 72 (29, p. 73). Therefore, zinc-72 would be the only isotope of zinc present in fallout fission products. With increased bombarding energy of the neutrons, a higher percentage of atoms of larger atomic number and atomic weight would be expected.

Evidence to date, however, seems to indicate that the amount of Zn^{65} in this area from fallout is inconsequential compared with that from the river. Davis et al (21) and Watson et al (71), working with organisms taken from the vicinity of the Columbia River, found much more Zn^{65} in terrestrial animals (fed forage irrigated with the river water) and marine organisms than did Murthy et al (47) in other areas of the United States. Lowman (44) stated that Zn^{65} only contributes about one-fifth as much radioactivity as natural K^{40} to Humbolt Bay, California, and Puget Sound, Washington, oysters. Seymour (65) agreed that organisms from Puget Sound were low in Zn^{65} activity, even though quite high in activity from fission products from fallout. This last work cited was carried out after the 1961 Russian test series.

The observations carried out during the course of this research tended to reaffirm the viewpoint that the contribution of fallout Zn^{65} is inconsequential. Figures 7 through 12 show the distribution of Zn^{65} off the Oregon coast, as evidenced by the radioanalyses of salps and euphausiids. Figures 7 through 9 were made from observations taken before the Russian test series, while Figures 10 through 12 were samples taken afterward. All values are picocuries per gram of dry weight. There appears to be no change in the Zn^{65} pattern that could be attributed to fallout. In fact, a comparison of the Zn^{65} pattern November 1961 (Figure 10), and the fallout pattern of Zr^{95}-Nb^{95}

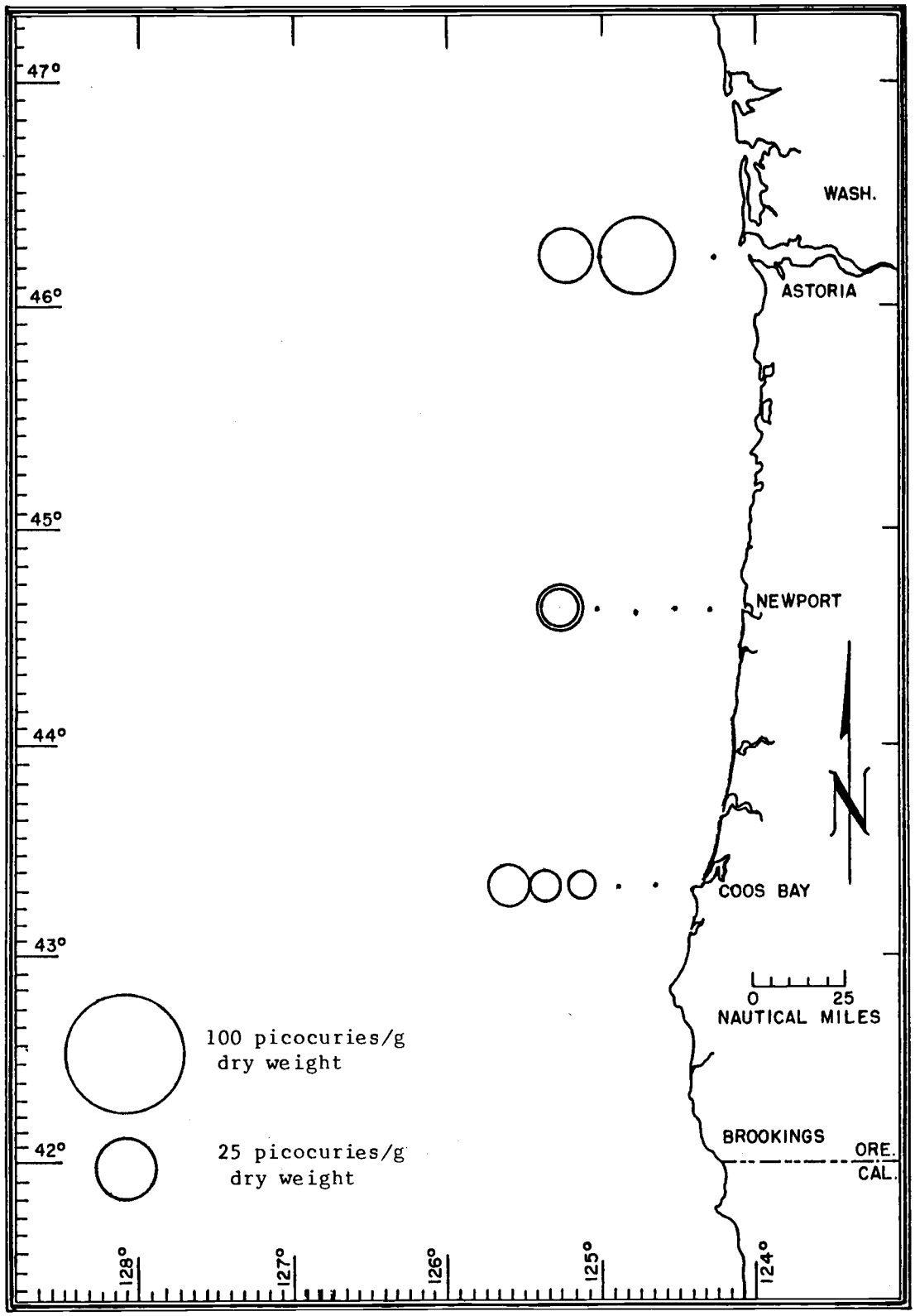

Figure 7. Distribution of Zinc-65 in salps, July 20-August 10, 1961

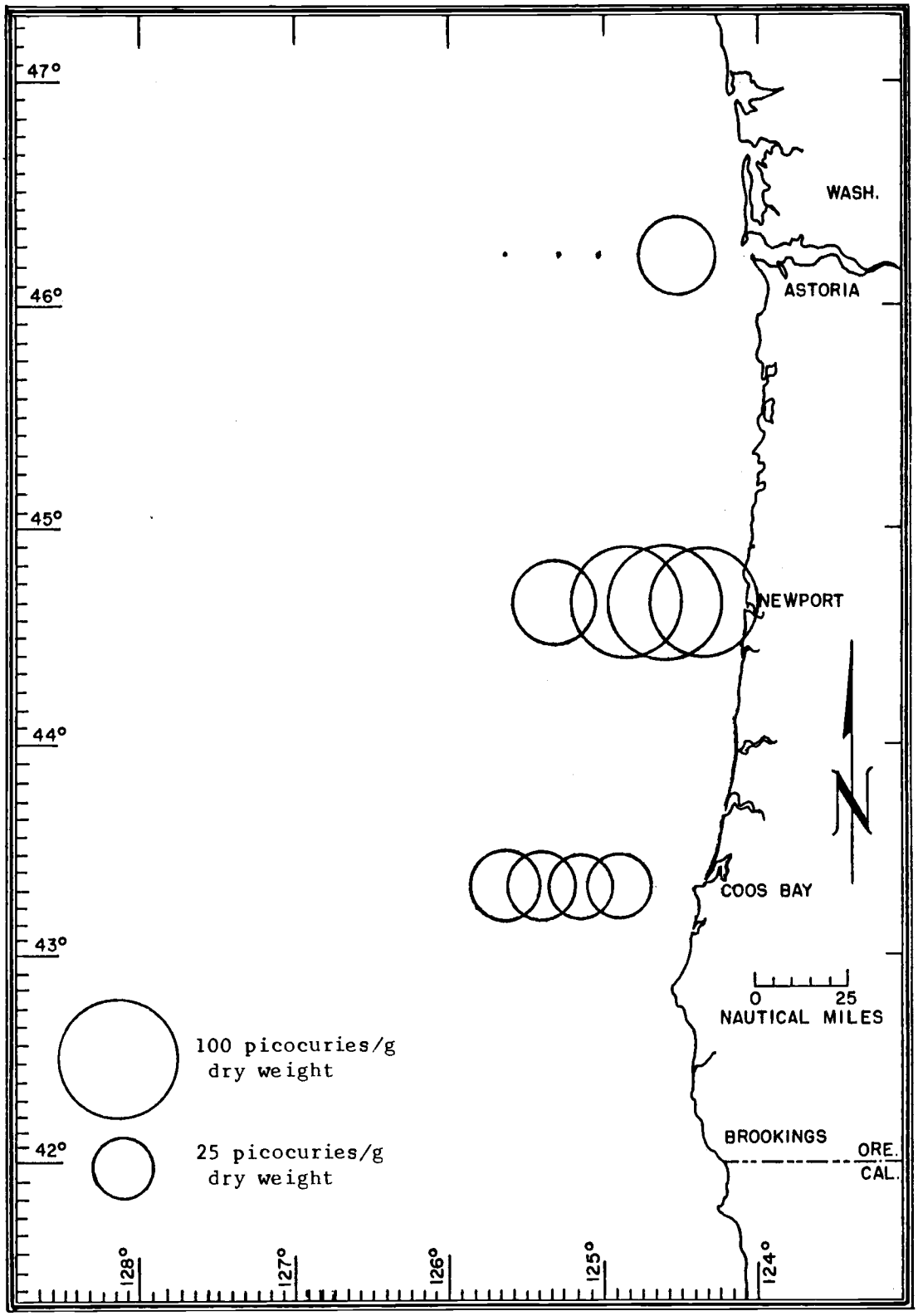

Figure 8. Distribution of Zinc-65 in euphausiids, July 20-August 10, 1961

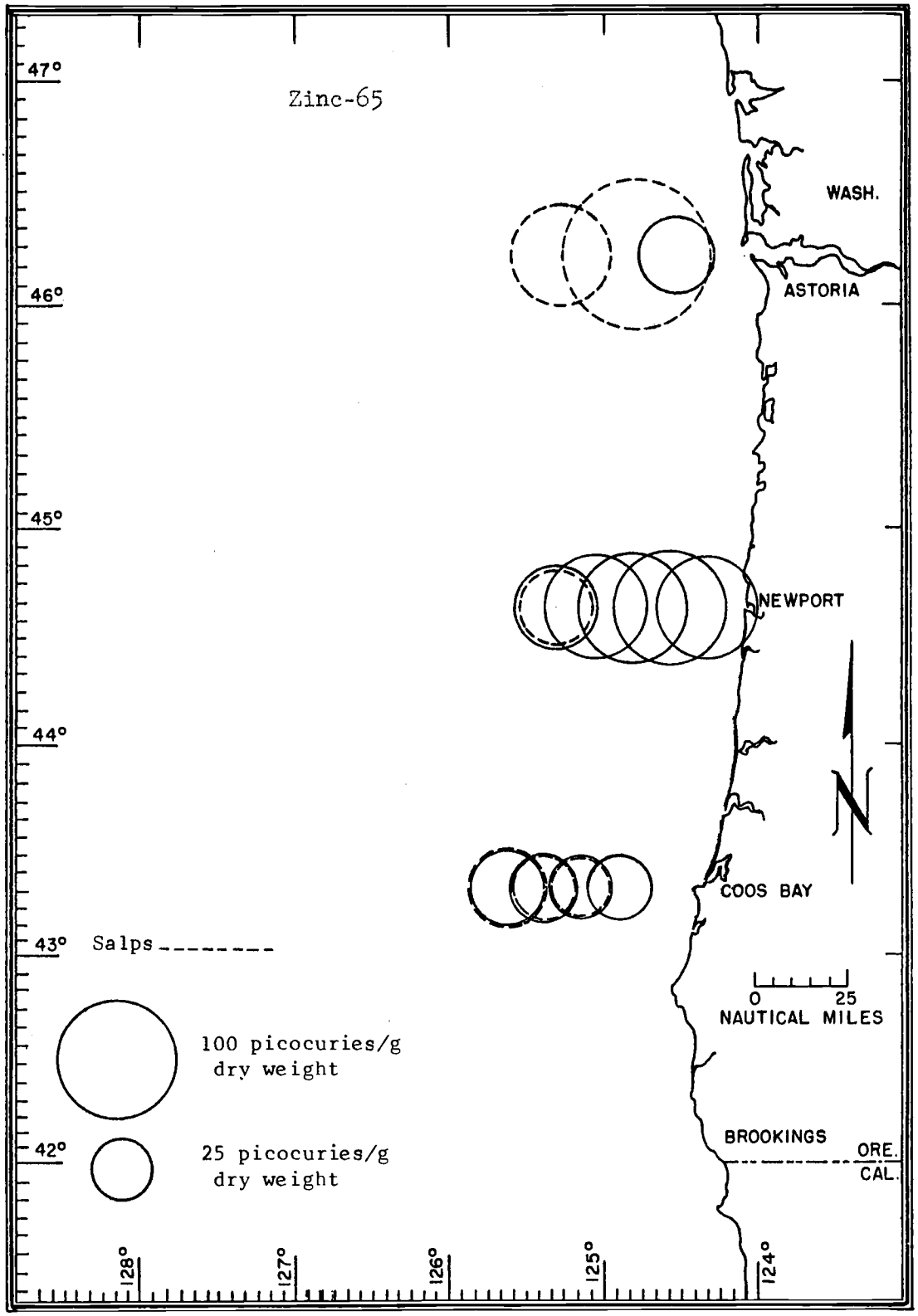

Figure 9. Combination of distribution of salps (X 3.6) and euphausiids, July 20-August 10, 1961

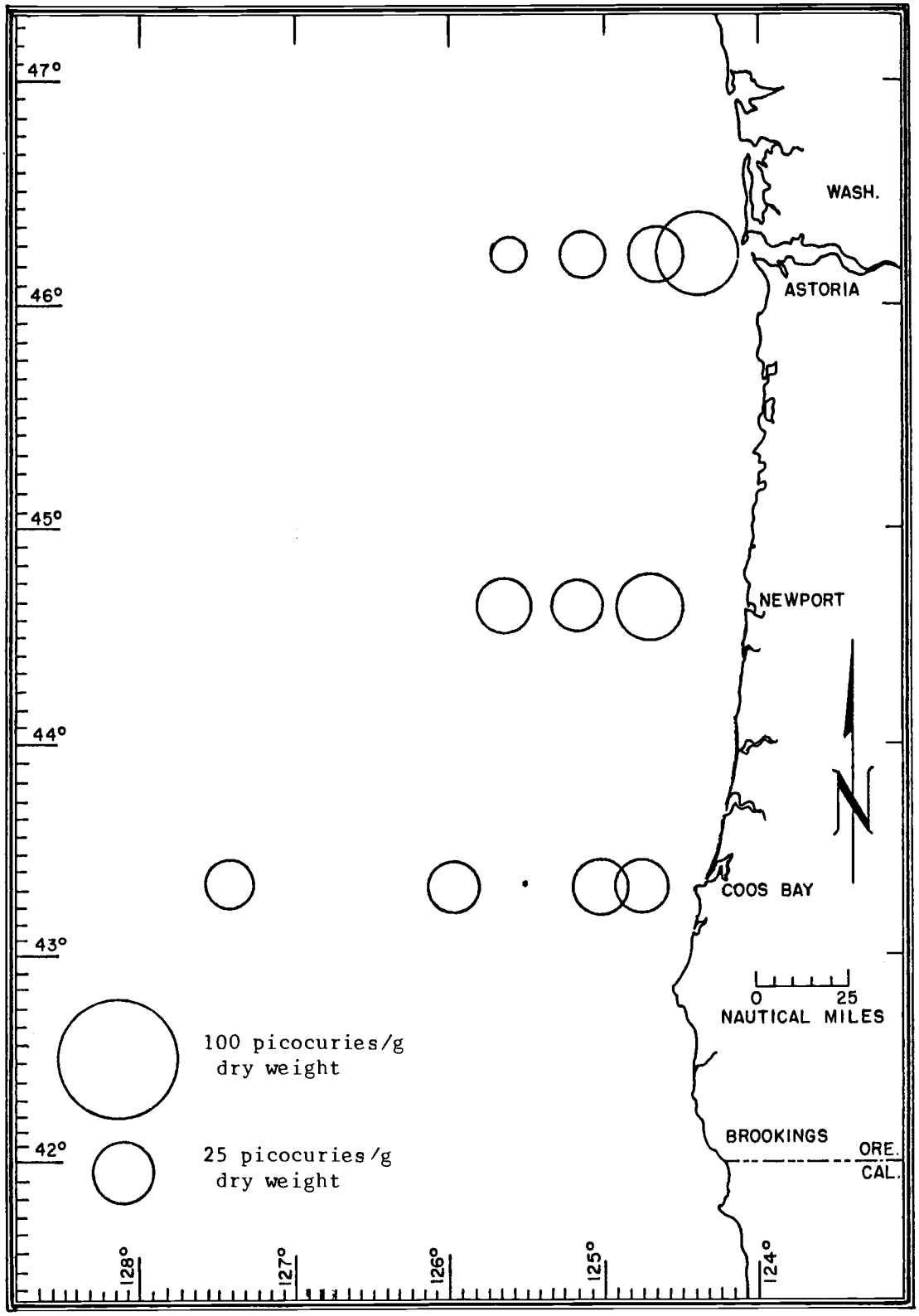

Figure 10. Distribution of Zinc-65 in euphausiids, November 6-15, 1961

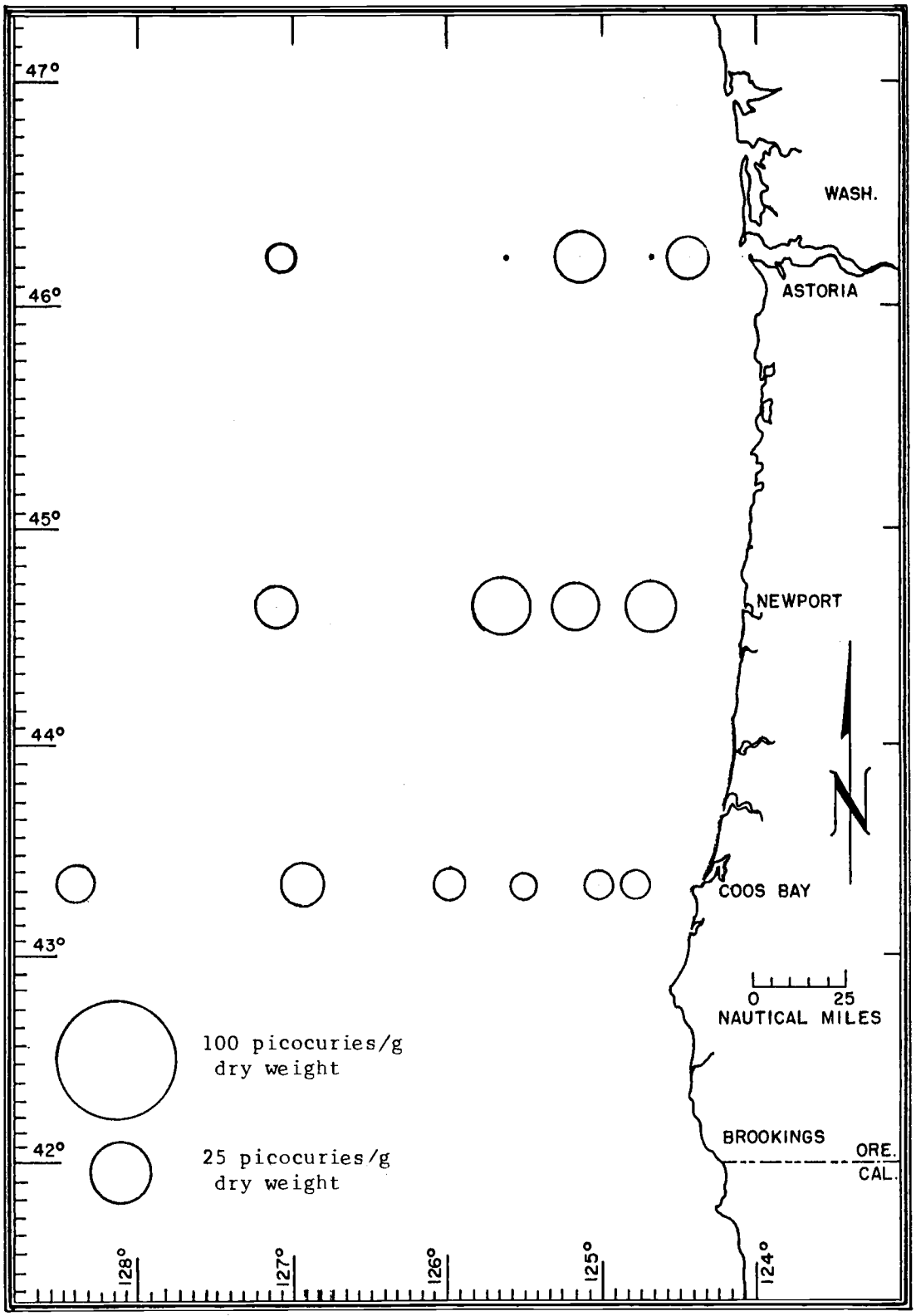

Figure 11. Distribution of Zinc-65 in euphausiids, January 8-16, 1962

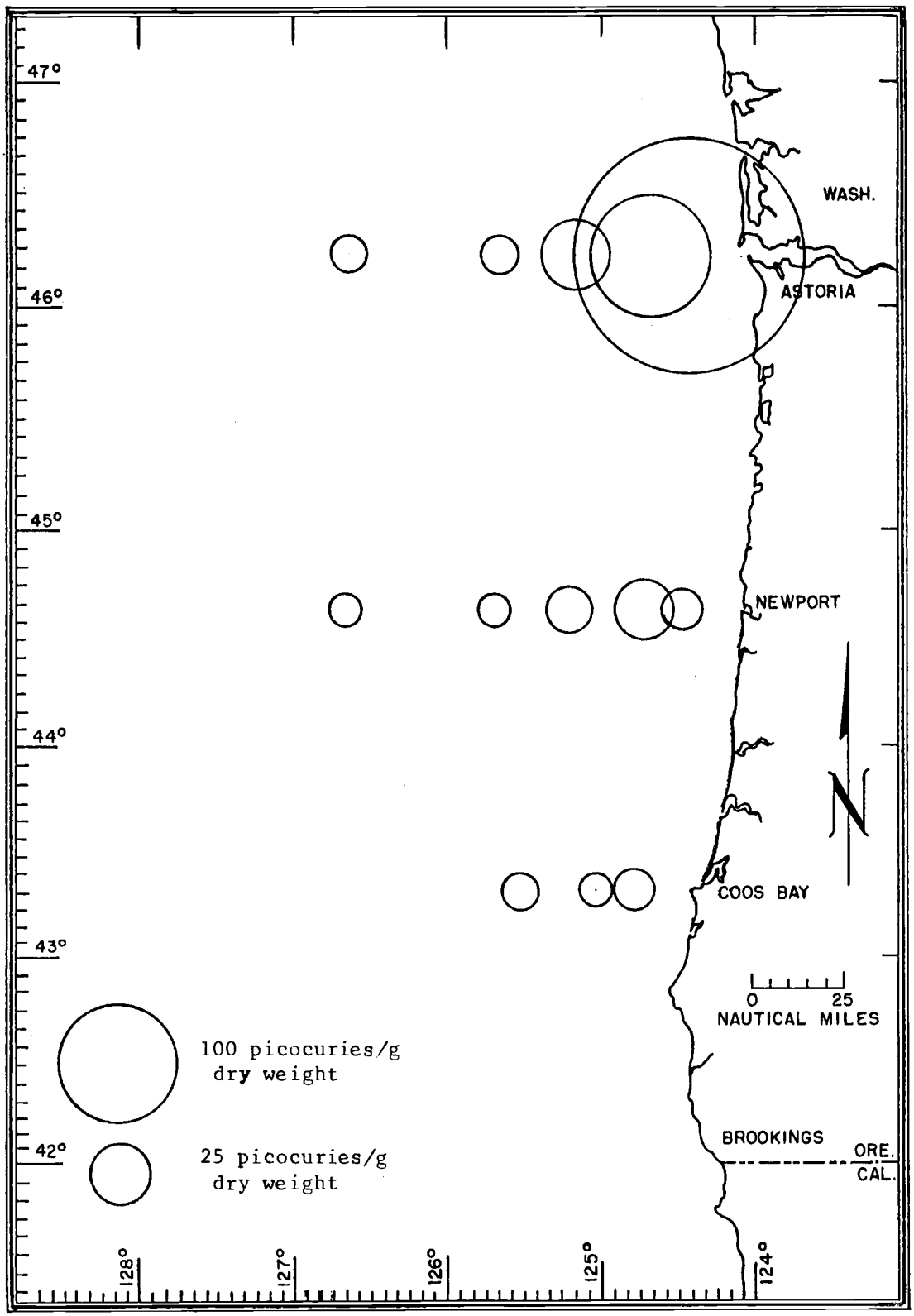

Figure 12. Distribution of Zinc-65 in euphausiids, March 26-April 5, 1962

(Figure 13) shows an inverse relationship near the mouth of the Columbia River, suggesting independent sources of the two radionuclides.

Further evidence was obtained from gamma spectra of euphausiids collected at much greater distances from the mouth of the Columbia River than could be provided by our routine trawl stations. Although only three such samples could be obtained, it is believed they were representative:

	DATE	LOCATION	ZINC-65 (per gram dry weight)
1.	July 23, 1961*	50°38'N, 178°06'W	1.0 ± 1.0 pc.
2.	Mar. 16, 1962**	34°19'N, 120°48'W	1.1 ± 1.0 "
3.	Mar. 18, 1962**	32°49'N, 123°54'W	1.4 ± 1.0 "

*Obtained from a U.S. Fish and Wildlife cruise, courtesy of Frank Hebard.

**Obtained from Dr. Edward Brinton, Scripps Institution of Oceanography, La Jolla, California.

The first sample, taken off the Aleutian Islands, was euphausiids collected before the Russian tests. The second and third, made off Los Angeles and San Diego, respectively, were of euphausiids collected after the nuclear tests. Though it appears there is a general low background due to Zn^{65}, it must be considered about an order of magnitude less than the amount found off the Oregon coast. The conclusion is that the Columbia River has played the dominant role in supplying Zn^{65} to the Oregon coastal region.

Before it can be stated with absolute certainty that Zn^{65} in euphausiids can be used to trace the Columbia River plume at sea, further work must be done on the retention time of zinc in euphausiids. Zinc-65 retention time has been measured in oysters, with rather strange results (60). It was found that the rate of accumulation of zinc from an environment containing Zn^{65} was greater than the rate of loss to a zinc-free environment. Furthermore, the loss of zinc from the oysters in a zinc-free aquarium environment leveled off after 40 days and remained stable for 60 days, but when the animals were put in a natural environment they rapidly lost more zinc in 30 days than they had lost in the previous 60 days. Also, the rate of loss was greater in the summer than in the winter.

It probably is not valid to extrapolate data on oysters in the laboratory to euphausiids in the open ocean, but it appears that euphausiids do accumulate Zn^{65} rapidly and lose it more slowly. For example, there seems to be no lag in the accumulation of Zn^{65} by euphausiids so that those nearest the mouth tend to contain the most. On the other hand, euphausiids at fairly great distances contain appreciable amounts of Zn^{65}, indicating some degree of retention (see Figures 8-12). Both

gain and loss of Zn^{65} may be accelerated in the summer. Superimposed on these seasonal effects, however, which appear to be related to metabolic activity, is the changing distribution of Zn^{65} in the ocean, with the whole picture confounded by a lack of knowledge of the effects of spring plankton blooms, diurnal migrations, etc.

Despite these difficulties, it should be pointed out that Zn^{65} appears to be at least as suitable a tracer of Columbia River water as is decreased salinity, and one that inherently possesses the capability of detection at much greater dilution. There is always the possibility that decreased salinity can be the result of rain storms, effluent from rivers other then the Columbia, etc. However those most experienced in tracing the plume of the Columbia River by salinity determinations appear confident of their techniques (5). Because of the concentration factors in plankton, the sensitivity inherent in the method, and the probability that measurable Zn^{65} is unique to the river, the radioanalysis of these ubiquitous organisms should prove to be a valuable additional tool in tracing the river at sea.

B. FISSION RADIONUCLIDES IN EUPHAUSIIDS

Since some of the fission products important in this study are produced both by nuclear detonations and, in trace amounts, by the Hanford reactors (48, p. 8), the problem of determining the origin of

the fission products in plankton arises. Barring an unreported accident of great magnitude, it is inconceivable that the escape of fission products from Hanford could account for the levels of fission products found in marine organisms after the Russian tests (Figures 13-21). Before the tests there was no evidence of any Zr^{95}-Nb^{95}, Ru^{103}, or Ce^{141} in the salps and euphausiids that were collected. The same sensitivity of analysis was used throughout the study. It therefore must be concluded that the amount of these fission products released by Hanford was below the level of detection, and the peaks that appeared in November were due to radioactive debris from the Russian tests of early September carried across the Pacific Ocean by atmospheric currents. Although ocean currents are known to transport radioactivity (46), the North Pacific Current is much too slow to have been responsible for this (50).

Particulate fission products were also found in euphausiids taken off Southern California after the Russian tests, though the radioactivity was lower than off the Oregon coast (50):

DATE & LOCATION	Zr^{95}-Nb^{95}	Ru^{103}	Ce^{141}
	(picocuries)		
Mar. 16, 1962 (off Los Angeles)	10.6 ± 1.4	0.2 ± 1.2	2.3 ± 1.4
Mar. 18, 1962 (off San Diego)	8.8 ± 1.3	0.2 ± 1.2	3.6 ± 1.4

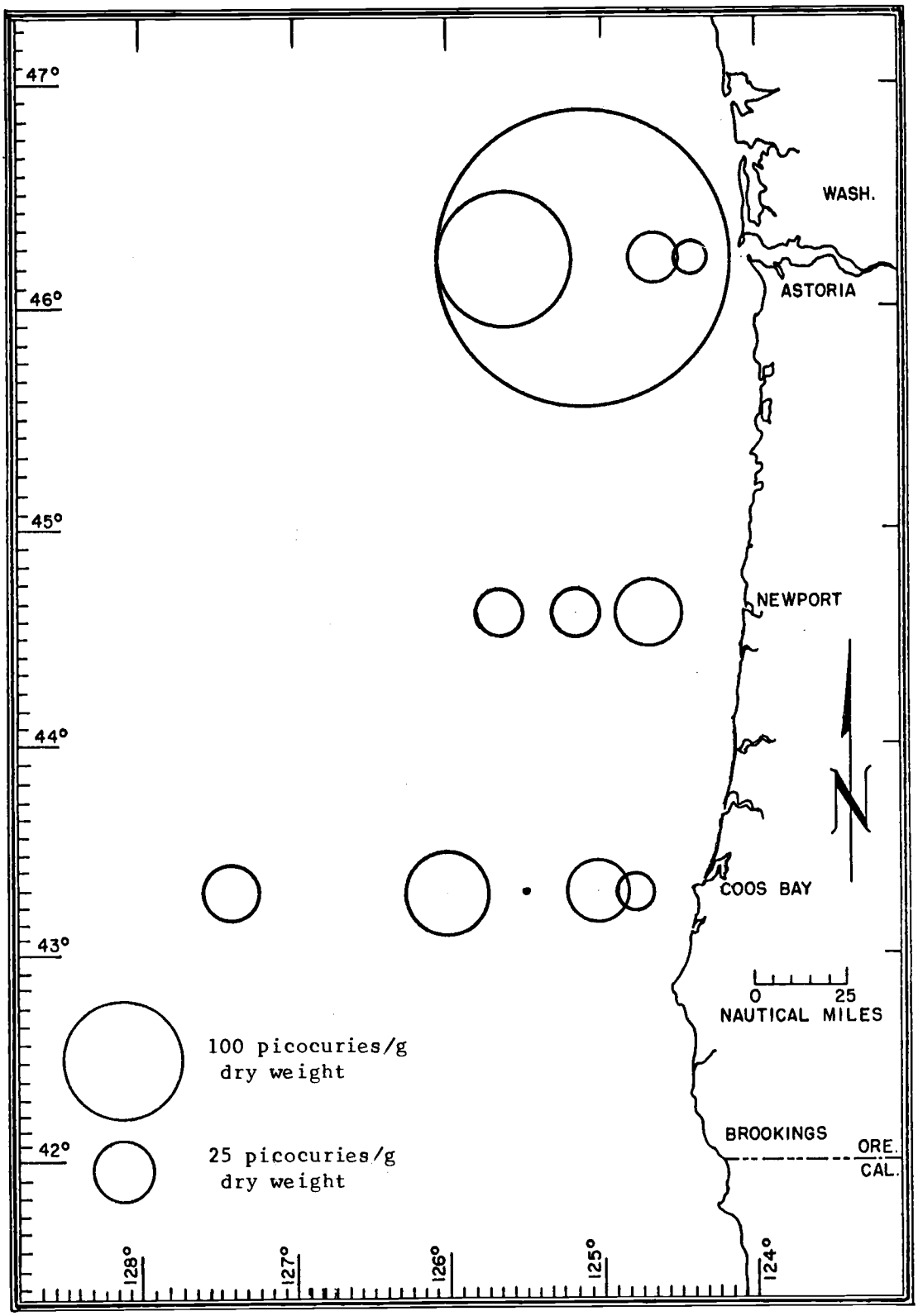

Figure 13. Distribution of Zr^{95}-Nb^{95} in euphausiids, November 6-15, 1961

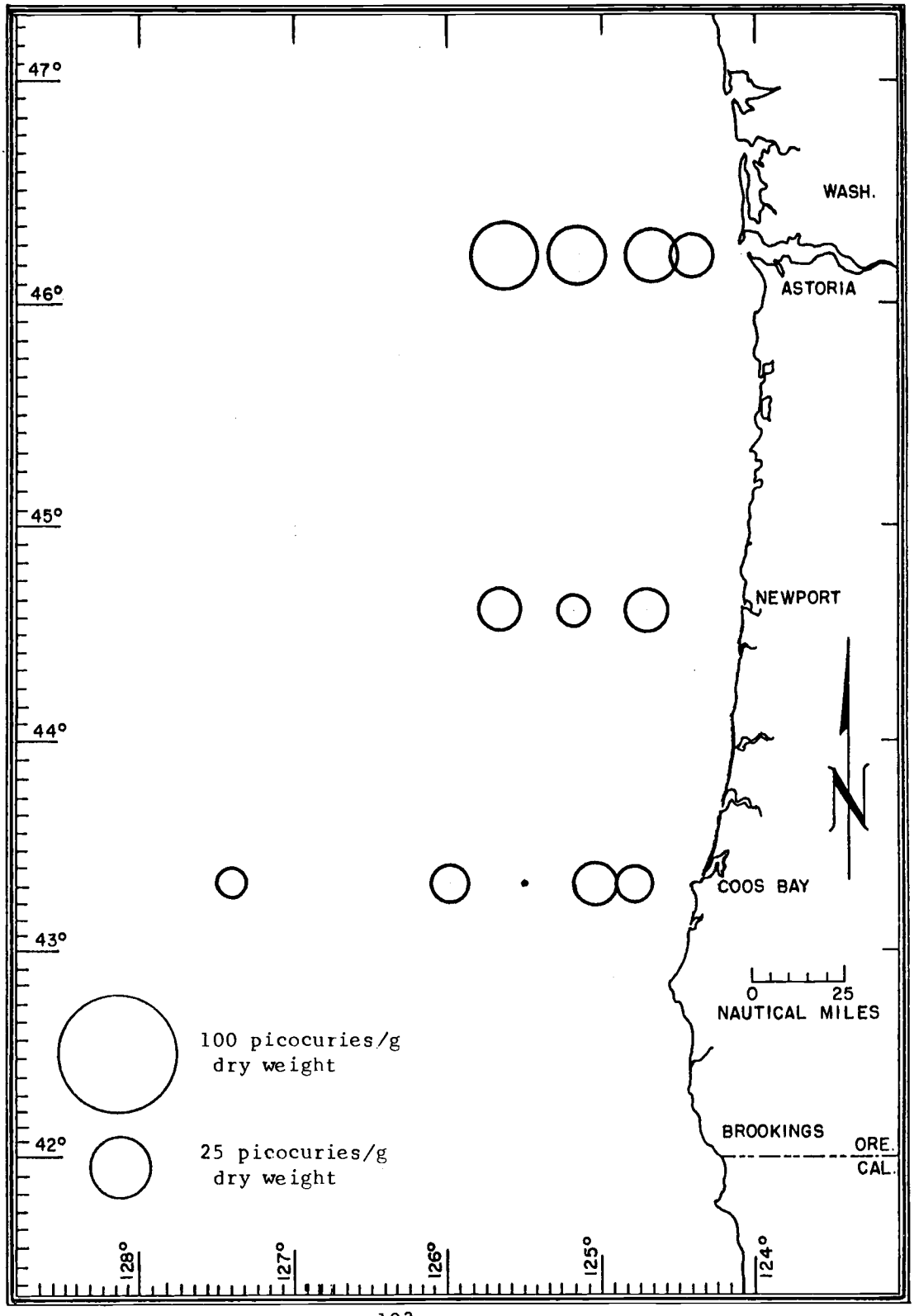

Figure 14. Distribution of Ru^{103} in euphausiids, November 6-15, 1961

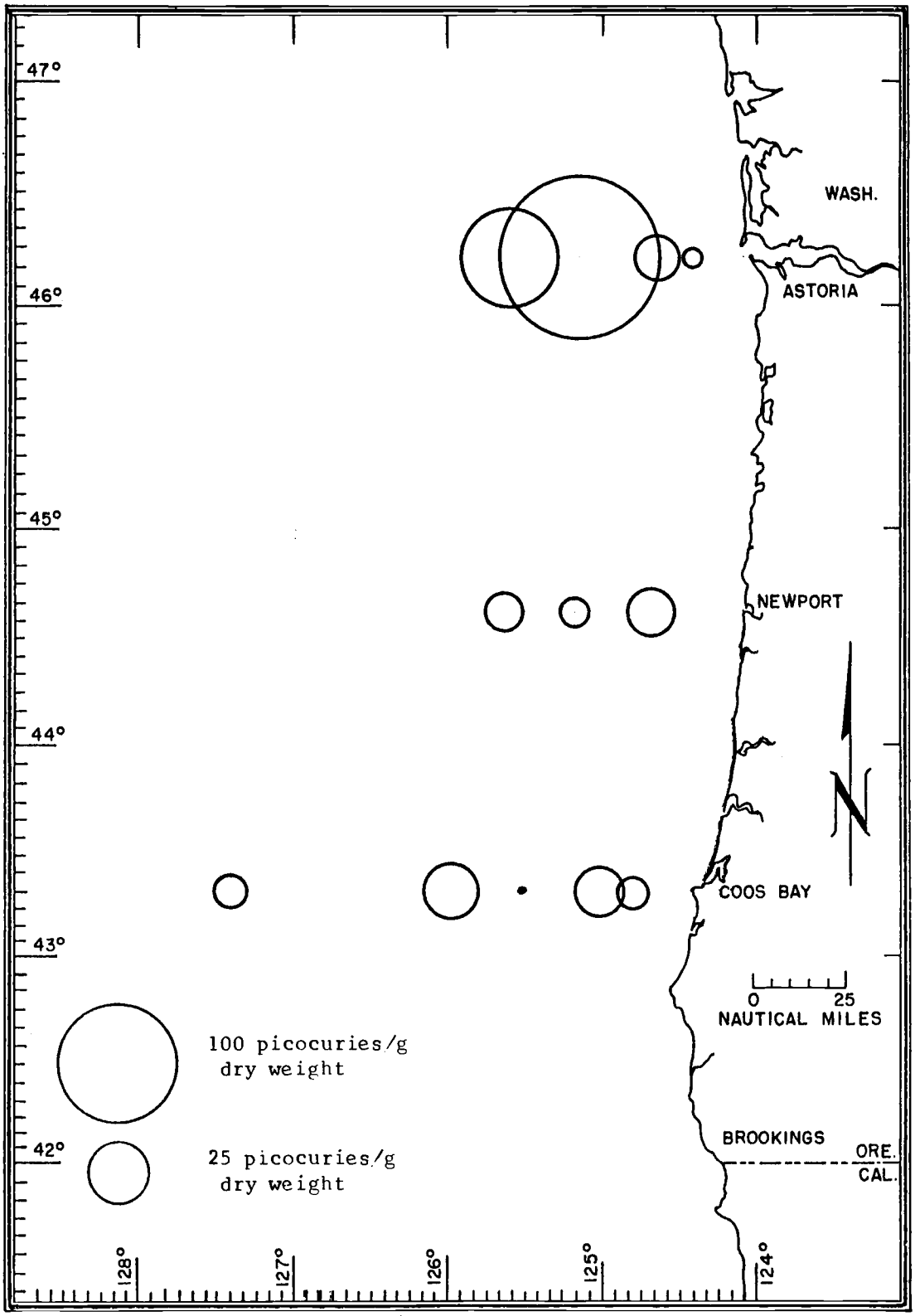

Figure 15. Distribution of Ce^{141} in euphausiids, November 6-15, 1961

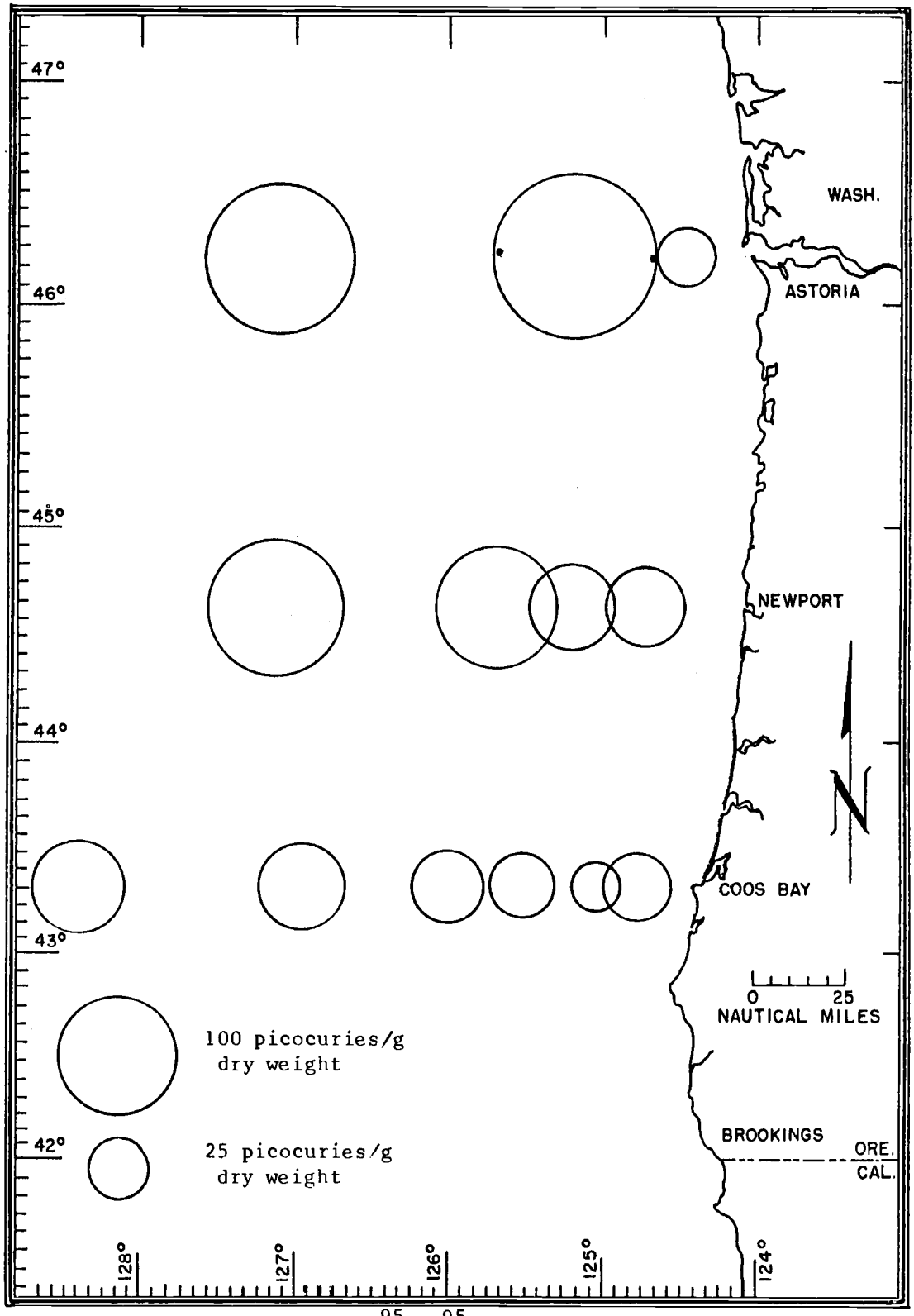

Figure 16. Distribution of Zr^{95}-Nb^{95} in euphausiids, January 8-16, 1962

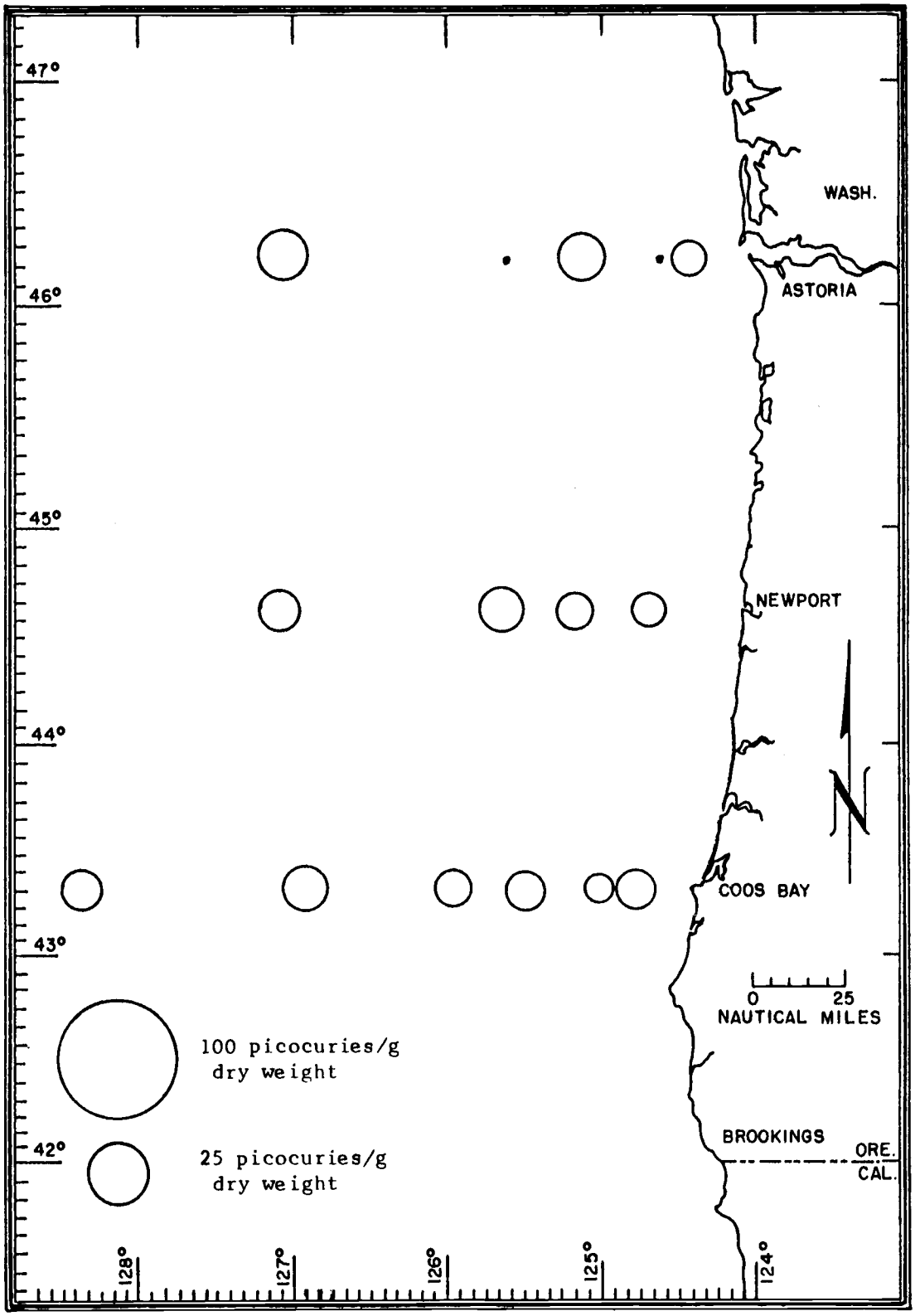

Figure 17. Distribution of Ru^{103} in euphausiids, January 8-16, 1962

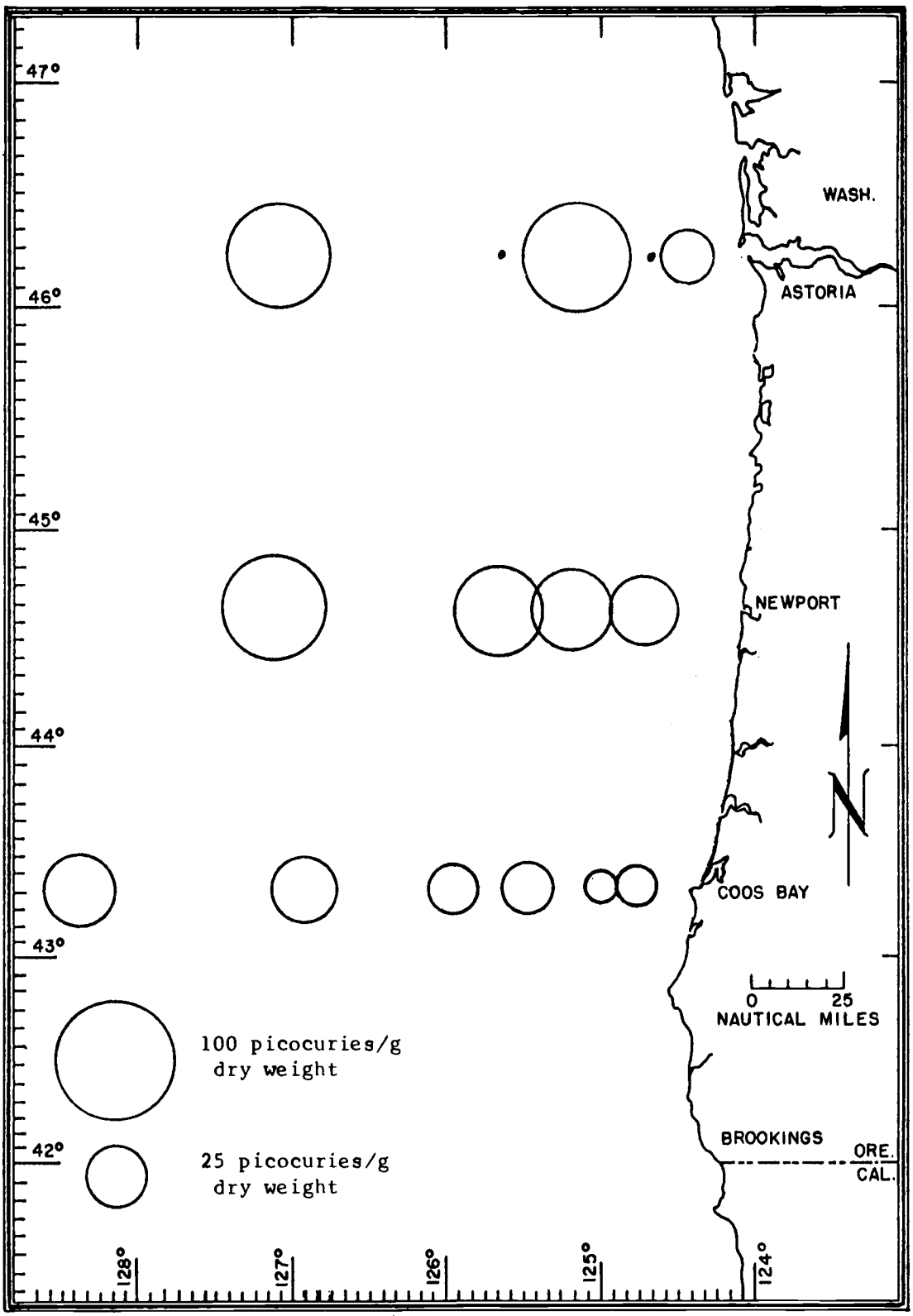

Figure 18. Distribution of Ce^{141} in euphausiids, January 8-16, 1962

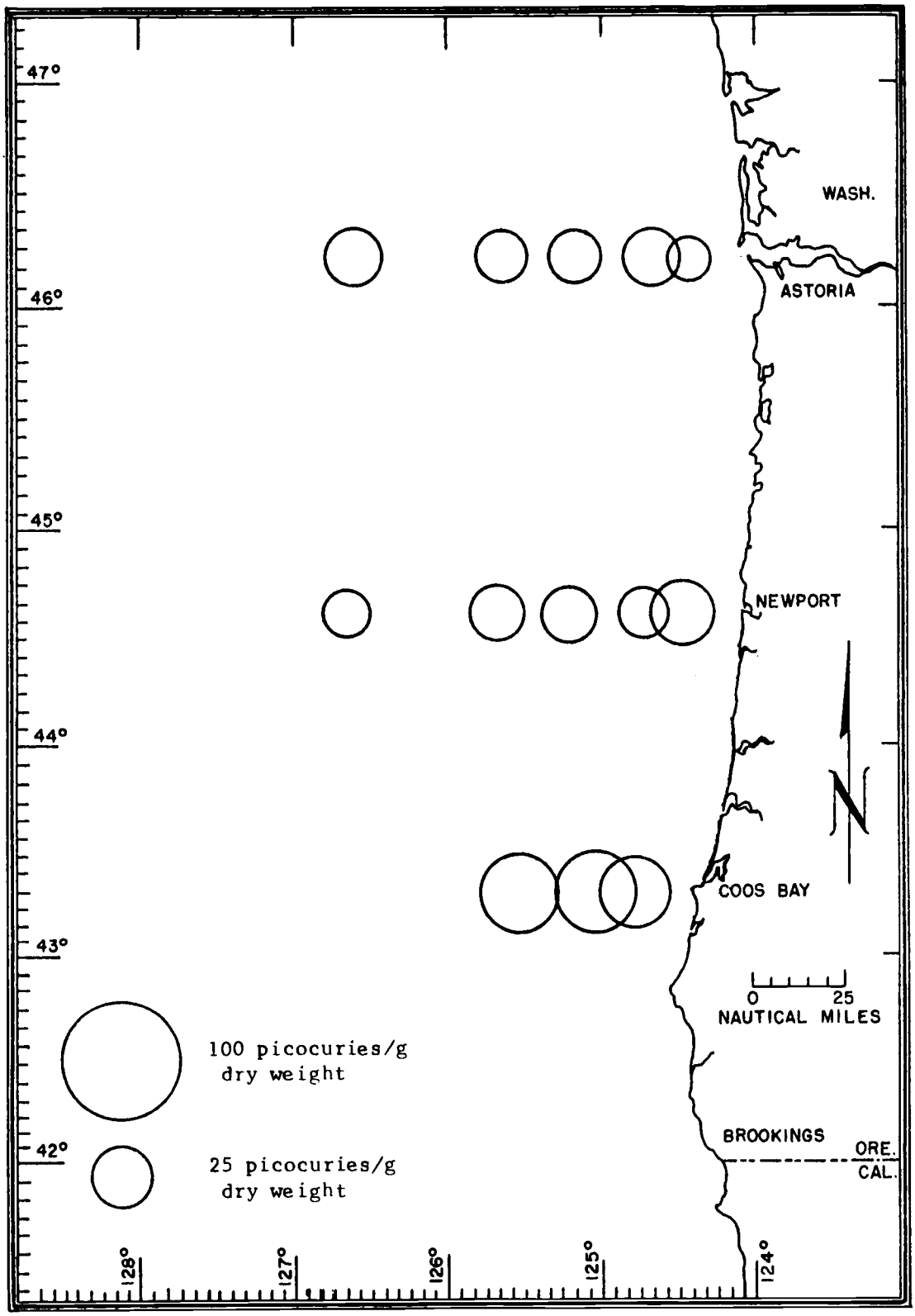

Figure 19. Distribution of Zr^{95}-Nb^{95} in euphausiids, March 26-April 5, 1962

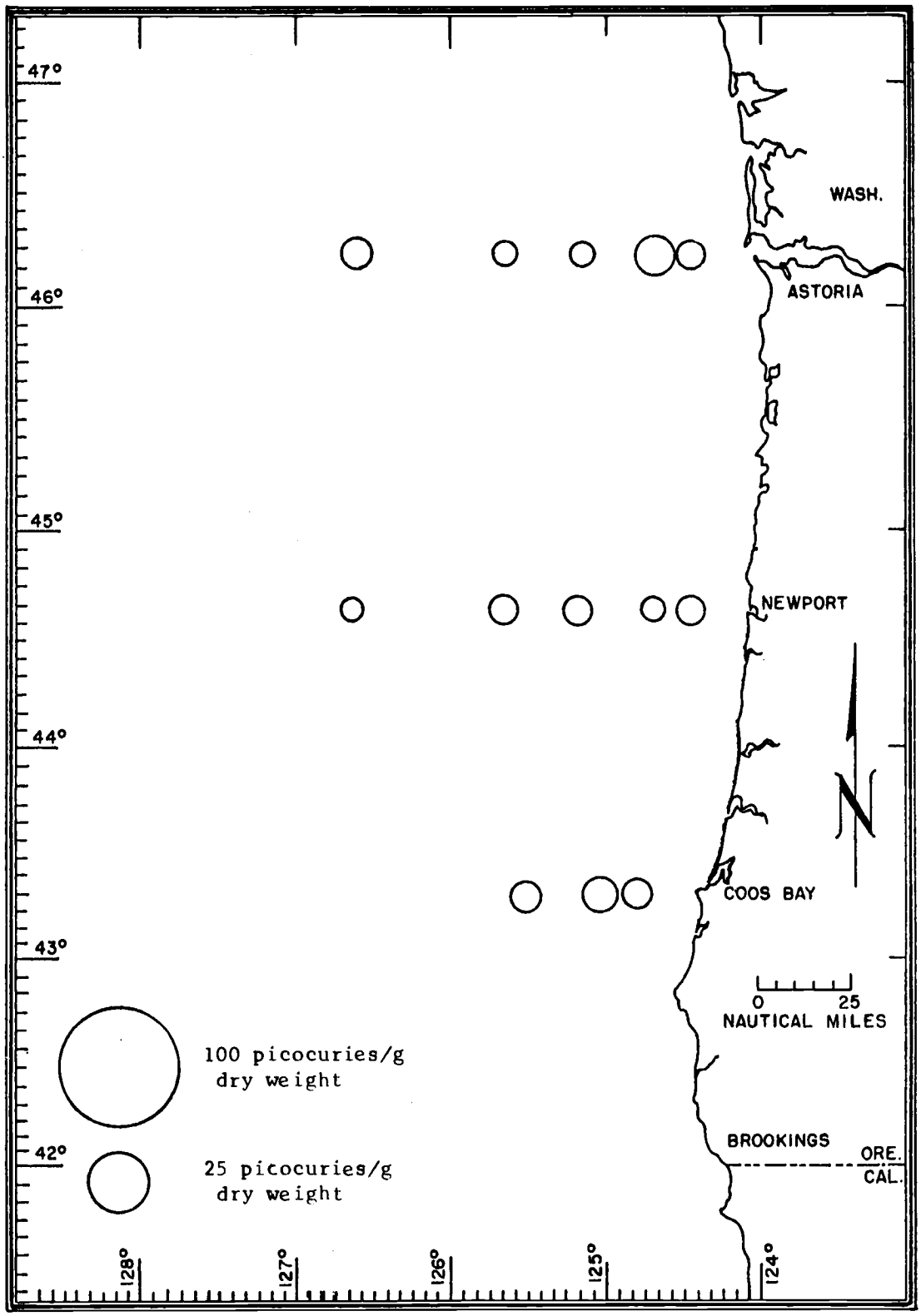

Figure 20. Distribution of Ru^{103} in euphausiids, March 26-April 5, 1962

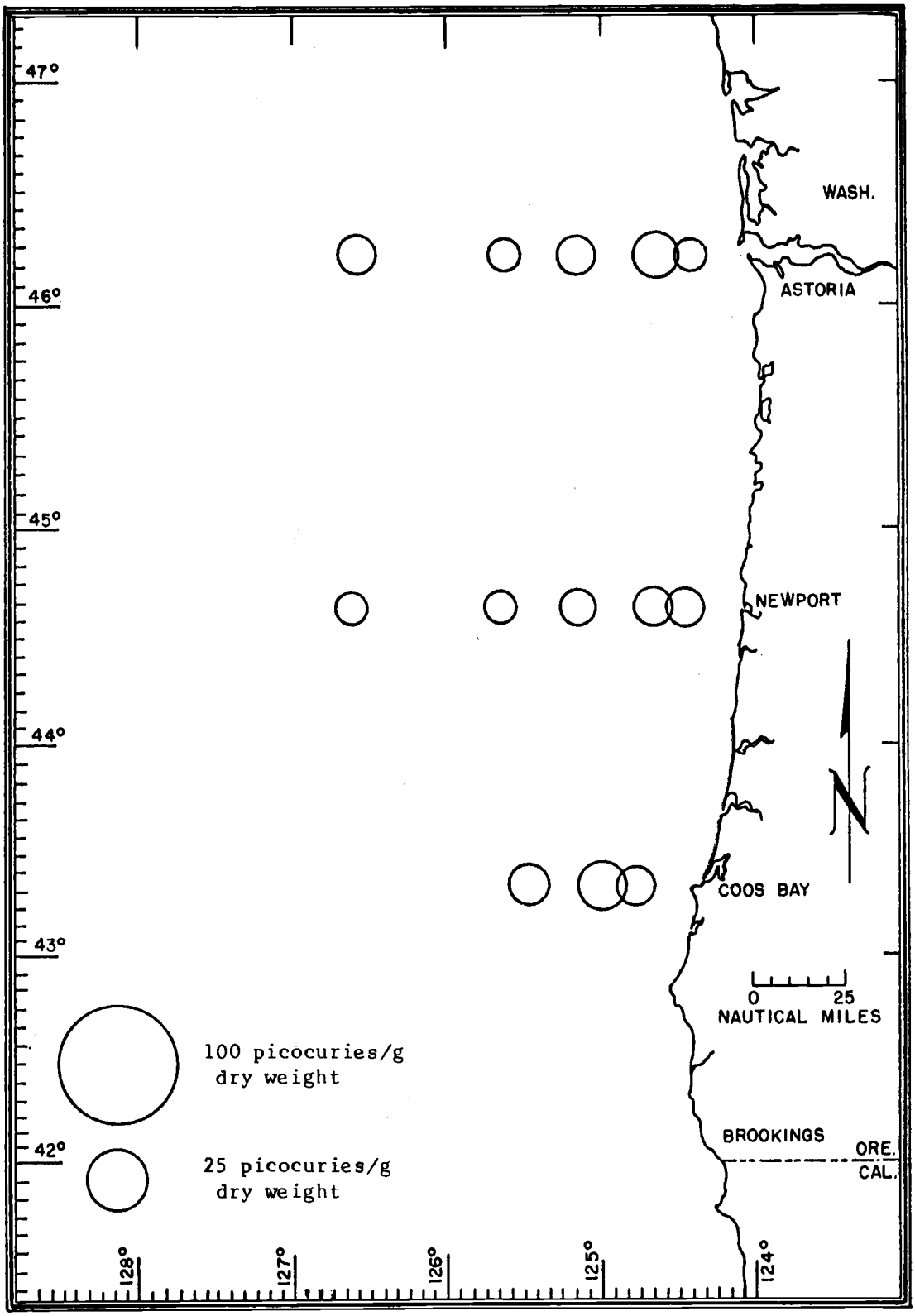

Figure 21. Distribution of Ce^{141} in euphausiids, March 26-April 5, 1962

The lower radioactivity (compare with Figures 19, 20, and 21, which incorporate samples taken at about the same time) was probably due to a latitudinal effect in the fallout pattern, and possibly also to irregularities in the fallout which are mentioned by Bowen et al (13) and Clarke (18).

After the Russian tests, it seemed advisable to analyze the radioactivity in the particulate matter off the Oregon coast on a transect out from the mouth of the Columbia River. It was possible that the huge watershed drained by the Columbia was acting as a "catch-basin" for a large amount of fission products. If these were washed into the river, they might make the river richer in fallout radioactivity than the ocean. Consequently, euphausiids and other filter feeders in the Columbia River plume in the ocean theoretically could be concentrating particulate fallout products that had been pre-concentrated by the river before being dumped into the sea, as well as those that fell directly into the sea. To test this possibility, surface sea water was filtered through a membrane filter, with glass fiber prefilter (see Materials and Methods). Only one series of measurements was made, and the accuracy of the experiment probably was poor because of small sample size and low radioactivity. Nevertheless, a graph of radioactivity versus distance from the mouth of the Columbia River shows a definite trend toward an

increase in the amounts of all measureable radioisotopes on the filters, both fission and neutron-induced, in the vicinity of the mouth of the river (Figure 22). Logarithms were used so that chromium-51 could be graphed on the same scale as the other less abundant isotopes, and also to smooth out some of the variations due to small sample size.

Zn^{65} and Cr^{51}, since they are produced at Hanford and normally appear in the river, increase on the filters with proximity to the mouth of the river as expected. Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141}, being present in fallout, are more difficult to evaluate. Superficially it appears that the river may be acting as a reservoir for these radionuclides, but circumstantial evidence indicates that this is not the reason for the increase of particulate fallout radioactivity on the filter in the proximity of the river. Part of this evidence is based on the fact that a similar increase in natural potassium-40 is suggested in Figure 22, when it is known that the sea is richer in K^{40} than the river (70, p. 10-12). Isotopes that are trapped on a filter must either be particulate or associated with particles in the water. Particulate material, both organic and inorganic, is more abundant near the shore. These particles must play a role in making the radioisotopes more available to the filter. Either the particles retain the radioisotopes present in the surface water used in the filtration experiment, making the water near the river appear more radioactive or they bind the radioactivity into particulate

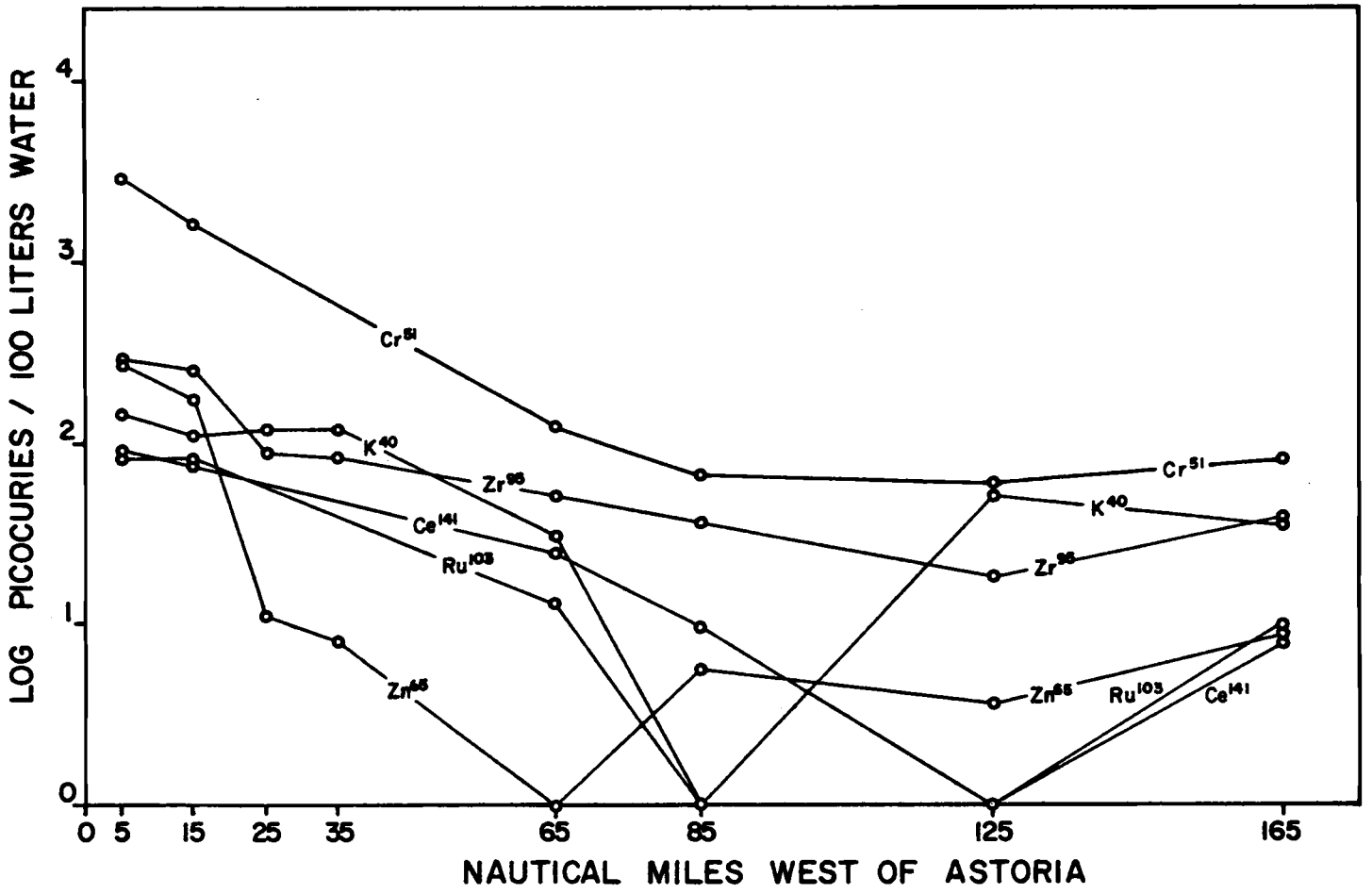

Figure 22. Radioactivity of filtered sea water taken on a transect west of Astoria, Oregon

matter that will be retained on the filter. This latter explanation seems valid for K^{40} or Zn^{65}, which , being ionic, would normally be expected to pass through the filter, but less valid for Zr^{95}-Nb^{95}, which are considered to be particulate in sea water. Since both chlorophyll a (Figure 23) and inorganic detritus increase toward the mouth of the river, it is extremely difficult to determine whether organic or inorganic material is responsible for the effect observed.

In addition to the behavior of K^{40} previously noted, additional evidence that the level of fission product radioactivity is not actually greater in the proximity of the Columbia River is shown in Figure 24, which is a plot of the radioactivity of euphausiids versus distance from the mouth of the river. The same radioisotopes are evaluated, this time on the basis of a gram dry weight of euphausiids rather than per volume of filtered sea water. The euphausiid samples, although collected within 24 hours of the filtered water samples, show great differences from the filters. Only Zn^{65} behaves the same way, increasing toward Astoria. All three fission products and K^{40} are more constant in the euphausiids, though they show a small peak 25 miles from Astoria (AH-25), and a sharp drop 10 miles nearer to shore (AH-15). Interpretation is difficult, but the drop at AH-15 may be significant, especially since it also was present in Figures 13-21. The behavior of Cr^{51} is different. Despite its abundance on the filter at AH-15 (Fig. 22), it

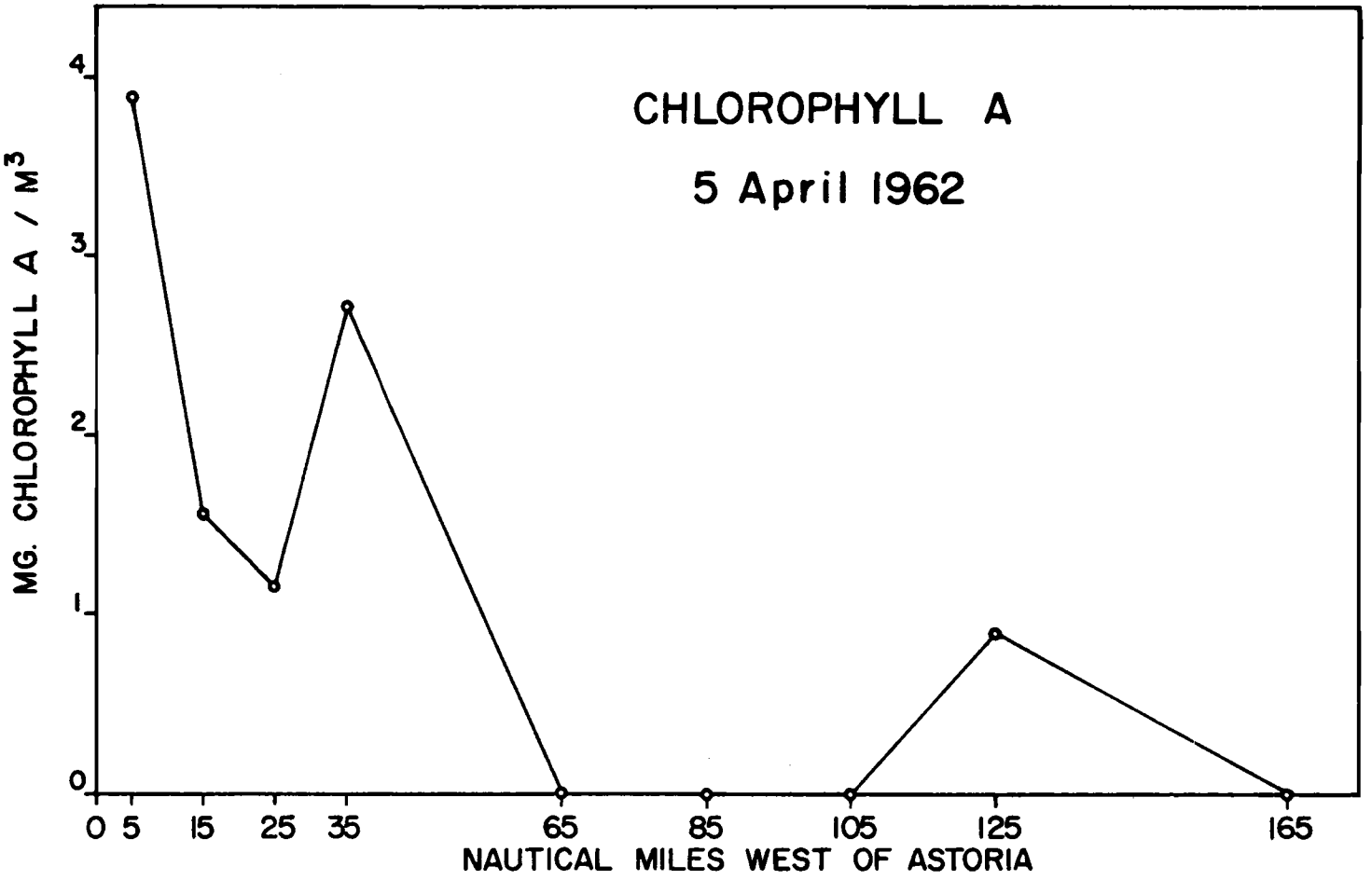

Figure 23. Chlorophyll a in sea water taken on a transect west of Astoria

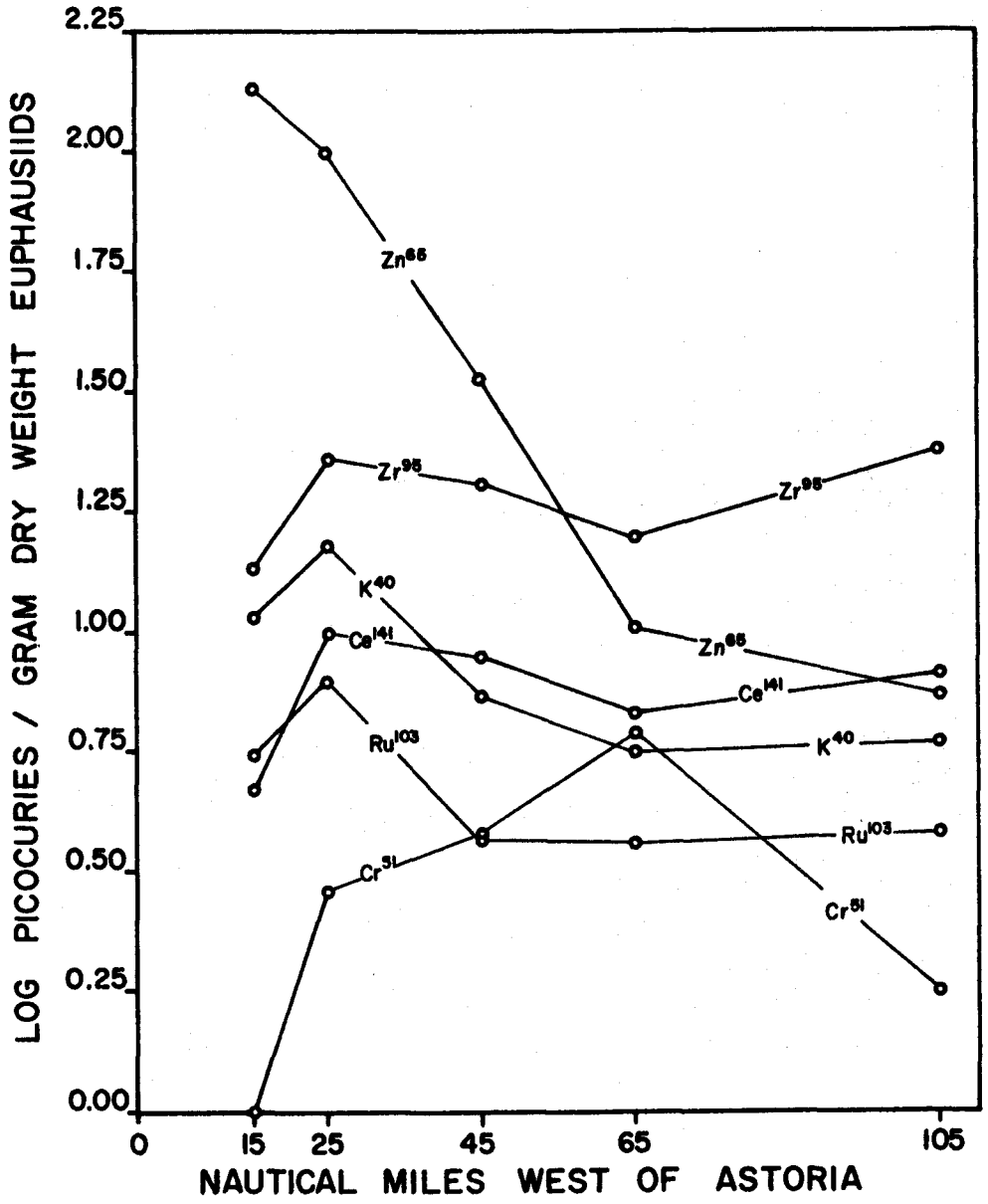

Figure 24. Radioactivity of euphausiids taken on a transect west of Astoria

is not detectable in euphausiids taken at that location (Fig. 24). Instead, in the euphausiids it exhibits a gradual rise going away from the river, reaching a maximum at AH-65. The accuracy of the analysis of Cr^{51} is very poor because its photopeak falls in a region which contains the Compton scattering from many other isotopes (see section IV, B). This might tend to invalidate the small peak observed in euphausiids taken 65 miles off Astoria, but certainly the large quantities of Cr^{51} found on the filters in the mouth of the river near Astoria are highly significant (note log scale!).

C. Horizontal and Vertical Transport of Radionuclides in the Sea

Occurrence of great levels of particulate fission products off the Oregon coast after the Russian tests afforded the opportunity to follow the dispersion of these products with time. Figures 13 through 21 trace the lateral dispersion off the Oregon coast, using Euphausia Pacifica as a monitor. Ocean currents probably played the major role in smoothing out the "hot spot" (Zr^{95} and Nb^{95}) of November, 1961 (Figure 13) into the less intense spot of January 8, 1962 (Figure 16) and finally into the much more uniform pattern seen in March-April, 1962 (Figure 19). Some influence must be attributed to the organisms themselves because of their habits, however. Euphausiids generally rise to the surface at night to feed on phytoplankton, with maximum feeding at about 1 a.m.

(55), and drop down to much greater depths in the daytime (22). These diurnal vertical migrations may carry them through layers of water that are moving in different directions laterally; for example, the animals may spend the hours of darkness in surface waters that are moving south, and the daylight hours in deeper waters that are moving north. Hardy shows some of the possible effects of vertical migrations on horizontal distributions (32, p. 213). Because of the uncertainties in the extent of the vertical migrations, and the vagaries of the water currents at all appropriate depths, little can be said of the overall effect of lateral transport in this area of the ocean, however. The only conclusion that seems certain is that the combined effect of vertical migration and ocean currents is one of dispersion.

Vertical transport of radionuclides is perhaps of more concern than lateral transport. The ocean is not a homogenous body of water, but rather it is poorly mixed so that there is a general decrease in temperature with depth. The condition of cold, denser water underneath lighter, warmer is a relatively stable one, and therefore very little vertical movement of water occurs. Molecular diffusion is negligible, and eddy diffusion, though much more important, is still extremely slow. Because of this, it has been assumed that radioactive materials dumped on the bottom in deeper sections of the ocean would be isolated sufficiently from any contact with man. Recent work, however, has led to some reevaluations of the residence time of water in the deep basins,

and the ocean now looks less desirable as a receptacle for atomic wastes (9). The Russians, in particular, have been strongly opposed to atomic "dumping" in the oceans (10).

The reverse process, the movement of radioactivity from the surface down through the depths, is slow for the same reason; that is, the stability of the water due to its stratification. The most critical region in the water column with regard to vertical transport is the pycnocline (usually thermocline). The thermocline is a transition zone between cold dense water and the warmer surface water, and acts as a barrier to most of the physical vertical transport. The water above the thermocline is generally well mixed, so that materials introduced in the surface will quickly spread throughout this whole zone (72). Lowman (43, p. 114) mildly disagrees, stating that the radioactive material was not uniformly distributed throughout the mixed layer in the neighborhood of the Marshall Islands, even though samples were taken 6-8 weeks after contamination of the water mass. In general, however, the layer above the thermocline must be considered relatively well mixed, with the thermocline preventing the extension of mixing further down.

If the radioactivity is confined to a particle, it could be transported downward across the thermocline by gravity. This process would be faster for large (heavy) particles, and in the absence of complications, Stokes's law would govern its descent. Conditions in the mixed

layer complicate the computed rate of descent of particles, however. Because of exposed polar groups, clumping can occur. While this might seem to speed the settling out process, the opposite result is possible. If the particle is adsorbed by phytoplankton (which have tremendous surface areas), it may be kept in the mixed layer by the buoyancy of the organisms. On the other hand, if coalescence occurs and creates a particle large enough, it may be removed by the many filter feeders (i.e. most copepods, salps, euphausiids, etc.) in the upper layers. The fate of a particle in the gut of a filter feeder is still not resolved. It may be released in a fecal pellet where it can be recycled by phytoplankton, or the organism may be devoured, etc. While the processes and possibilities are varied, the effect of living organisms in the upper layers seems to be to offset the effects of Stokes's law, at least for some of the radioisotopes. Sugihara et al (67), however, report that cerium is removed (from the upper layers of the ocean) on particles sinking at about 100 m/month or slower.

The nuclear weapons tests have facilitated the checking of physical, vertical transport from the surface. In one test, the lower boundary of radioactive water moved downward at about 0.1 cm/second until it reached the thermocline where it abruptly stopped (59, p. 13). The same test showed that the radioactive materials, when introduced at the surface, spread out to cover an area of about 100 square kilometers

while maintaining a thickness of the order of a few meters (59, p. 14). Clearly physical, vertical transport is greatly inhibited by the stratification of the ocean.

Biological transport might be an important means of vertical transport in some areas of the ocean. Many plankters are known to make vertical migrations which involve crossing the thermocline, and it seems possible that they could disperse much radioactivity in a fairly short time, in view of their ability to concentrate the radioactivity. Donaldson (23) concluded that biological activity is often of greater importance than physical factors in the distribution and localization of radioisotopes in the marine environment. Ketchum et al (37) stated that given a surface concentration of zero radioactivity initially and a 1 cc/m^3 population of organisms that vertically migrated 100 m in one day, a concentration factor in the organisms of 340 would be needed to give a biological transport equivalent to the expected physical transport.*

*This mathematical relationship was presented by the senior author at the symposium, "The Biological Implications of Radioactive Isotopes in the Sea," Davis, California, June 19-22, 1961 (38). At the time of the presentation there was no quarrel with the concentration factor, which is very modest. It was felt by many that a population of 1 cc/m^3 was greatly overestimated, however. Lowman (43, p. 124) cites a survey by the Pacific Ocean Fisheries Investigation, which found 1 ml of plankton per 100,000 liters of water to be more representative of the open ocean. These estimates differ by two orders of magnitude but do not necessarily invalidate the conclusions.

Since concentration factors go up to 10^5 for some radionuclides in some organisms, the authors concluded that "it seems inevitable that the ultimate distribution of radioisotopes added to the deep sea would be profoundly influenced by biological processes."

So little is known of vertical migration, concentration factors of radionuclides by various genera in the ocean, and transport of radionuclides through food chains, however, that the supposition of great radioactive transport by biological means lacks universal support. Recent indications are that plankton only play a small role in the vertical transport of the total radioactivity. It is stated, for example, that strontium-90, cerium-144, and promethium-147 move in a non-biological system; i.e., their movement in the ocean is purely hydrodynamic (14). After the tests at Eniwetok, it was found that even though the radioactivity per unit volume of plankton was much greater than that for an equal volume of water, the average total radioactivity of the water was 40,000 times that contained in the micro-macroplankton (43, p. 136). Plankton clearly accumulate some radionuclides, but it would appear that their biomass is insignificant. This may be generally true if one considers the entire ocean, but perhaps is less true in coastal waters. It is also well known that equatorial oceans, where the Eniwetok work was done, are notoriously poor in net plankton compared with the temperate oceans. Such facts tend to suggest that plankton in

temperate, coastal waters, such as off the Oregon coast, cannot be ruled out as possibly important agents in the transport of radionuclides.

Even if plankton do fail to play a major role in the vertical transport of "total" radioactivity, this does not preclude an important role in the transport of a specific radionuclide. Living organisms making migrations would most effectively transport readily labile materials, i.e., materials which reach equilibrium rapidly with the dissolved phase (37). Unfortunately, the lability of the various isotopes in macroplankton is not known. Zinc-65 is not labile in this sense, because it forms chelates and is tightly bound in the organism. In another sense it is extremely labile, however. In the present study, for example, the zinc-65 associated with the euphausiids appeared to be readily transferred to other organisms higher on the food chain. This is inferred because Chipman (17) has shown that in general radionuclides reach the higher trophic levels most efficiently through the food chain. Perhaps in this "food chain" sense Zn^{65} becomes an important part of the vertically transported radioactivity in the sea. Rice (60) states that zinc-65 will probably prove to be more important than any fission products as a source of radioactivity in the ocean.

Ommastrephes, a pelagic squid, often has been mentioned with respect to the vertical transport of radionuclides, because it is known to make vertical migrations of several hundred meters (37). The

organism chosen for the present study, Euphausia pacifica, is perhaps better suited for radionuclide transport studies, however. It is closely related to the copepods, which appear in swarms in the sea (comparable numerically with insects on land). Euphausiids are sufficiently abundant to study routinely and play fundamental roles as grazers of phytoplankton and as food for predators. While considered planktonic, the E. pacifica swims actively upward and downward, diurnally. Its rate of swimming is probably similar to that of its near relative, the euphausiid Meganyctiphanes norvegica, which in the laboratory has covered as much as 305 feet per hour vertically upward and 420 feet per hour downward. In short bursts, it can almost double these rates (32, p. 208).

Recent work on the deep scattering layers in the ocean (layers of living organisms which bounce echos back to depth finders, giving the illusion of a "phantom bottom") indicates the layers can move downward as much as 25 feet a minute. Euphausiids, sergestids, and lantern fish were presumed to be the organisms that made up the three layers observed by Dietz (22) off California. It is of note that these three groups are common in our midwater trawls, where they are captured mostly at night near the surface. Both lantern fish and sergestids (see figures in section V, D, p. 88) were subjected to radioanalysis with results indicating the possibility of some vertical transport of radionuclides as

a consequence of diurnal migrations. The principal role, however, must go to the euphausiids, which sometimes made up 95% of the biomass in the nets and were almost never absent from night-time trawls (see Appendix IV). Their great abundance (confirmed by over 230 midwater trawls off the Oregon coast) and radioactivity (as evidenced by nearly 100 analyses of different euphausiid samples by gamma-ray spectrometry) coupled with their diurnal migrations, must combine to make them a tremendously efficient agent in the vertical transport of radionuclides.

In an effort to check vertical transport of radionuclides, the radioactivity of a single sample of euphausiids taken at an approximate depth of 1000 meters (determined by total wire out and trolling speed, plus calibrations) was compared with that of a sample taken a day later near the surface at about the same location. The deeper sample contained considerable radioactivity (see Figure 25), most of which presumably was picked up while feeding near the surface. This would indicate substantial transport across the thermocline, which is invariably found nearer the surface. Unfortunately, not much significance can be attached to this observation because of the sampling gear used. The nets were not the closing variety, and therefore sampled all waters enroute to depth and back to the surface. The nets were actually at depth 71 minutes. Sixty-two minutes were required to reach depth, and 28 minutes were needed to reach the surface after the run, giving a

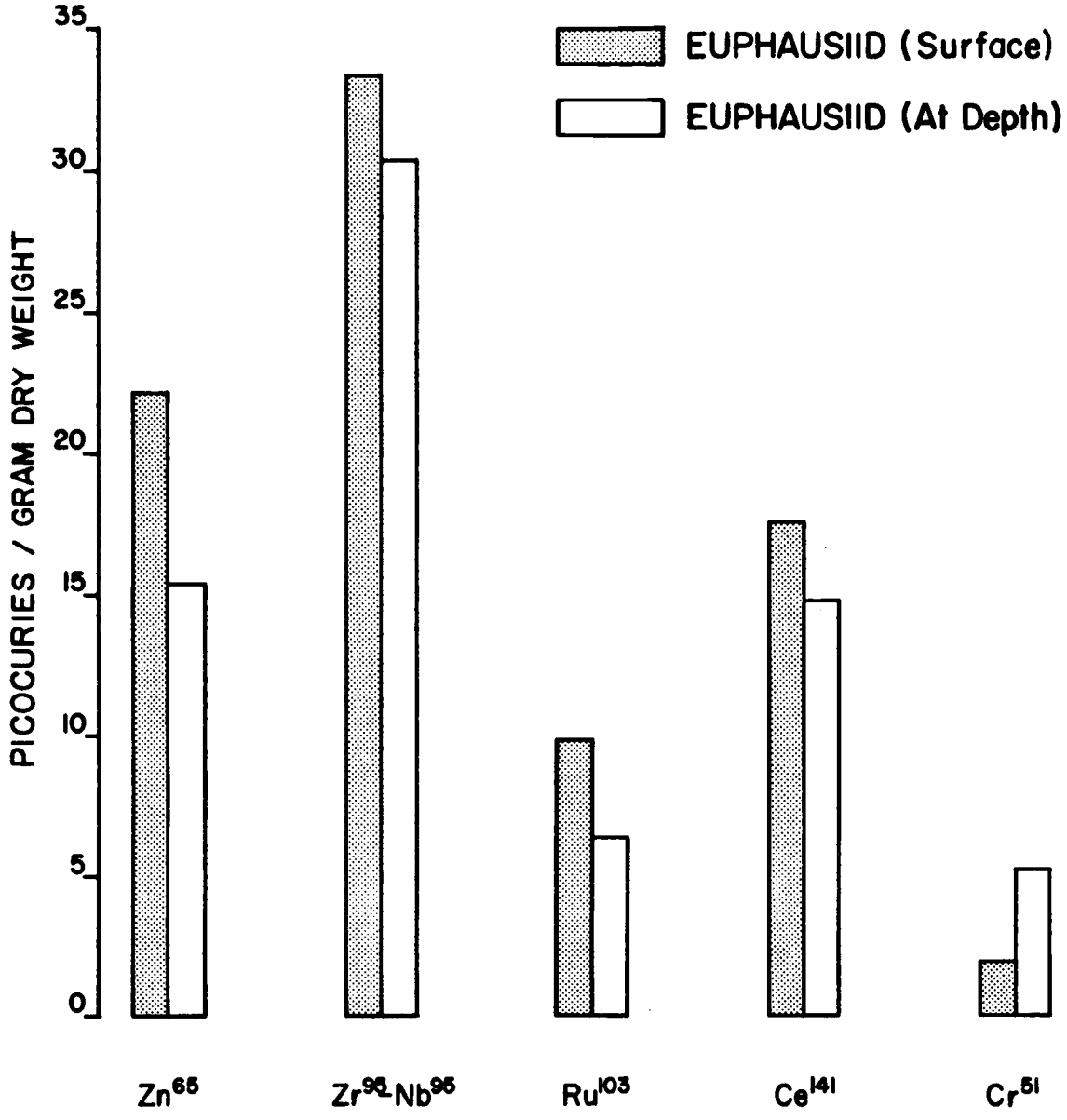

Figure 25. Comparison of radioactivity in euphausiids taken at surface and 1000 m.

total of 161 minutes in the water. The net fishes very inefficiently going down, but certainly obtains an integrated sample at all depths when coming to the surface; thus there is the possibility that no euphausiids were obtained at 1000 meters, but rather they were all taken in surface waters on the retrieve. The appearance of the surface euphausiids and deep water euphausiids was different, however, perhaps indicating a true difference between the groups. Those from the deep tow were somewhat larger and lighter in color than those from the surface. Only closing nets can obviate the argument.

D. The Passage of Radionuclides Through Several Trophic Levels

The Isaacs-Kidd midwater trawl sometimes collects several gallons of small marine organisms in a 30-minute tow. In hauls of this size to date, the bulk of the biomass usually has been euphausiids or salps. Often the hauls rich in euphausiids contained numbers of small bathypelagic fish. One of these, a lantern fish, Lampanyctus leucopsarus, feeds on euphausiids to a large extent as indicated by stomach analyses of the fish used for radioanalysis. This lantern fish therefore represents a third trophic level.

In addition to the second trophic level (euphausiids) and third trophic level organisms, one particularly bountiful haul made 15 miles off the mouth of the Columbia River contained many carid shrimp (mostly Pasiphaea pacifica, but some P. magna), which are probably omnivores. The stomach analyses indicated these particular specimens were almost entirely predaceous, although the stomach contents were well masticated, so that only occasional appendages were discernible. At the same station that yielded the numerous second and third trophic level crustaceans, surface water was filtered to measure the radioactivity in the phytoplankton and detritus in the water (first trophic level). Therefore, at this one location a measure was made of the relative amounts of radioactivity in three trophic levels. The results are shown in Figure 26. It can be seen that the levels of radioactivity in the carid shrimp most closely resemble those of the lantern fish, possibly indicating a similarity in diet.

Since Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141} are thought to be particulate in sea water (31)*, it is worth noting that they were found in the euphausiid, which is a particulate feeder. They were discriminated against at the third trophic level, appearing only in traces. In contrast

*A later reference (17) lists ruthenium as non-particulate in sea water but acknowledges that a question does exist.

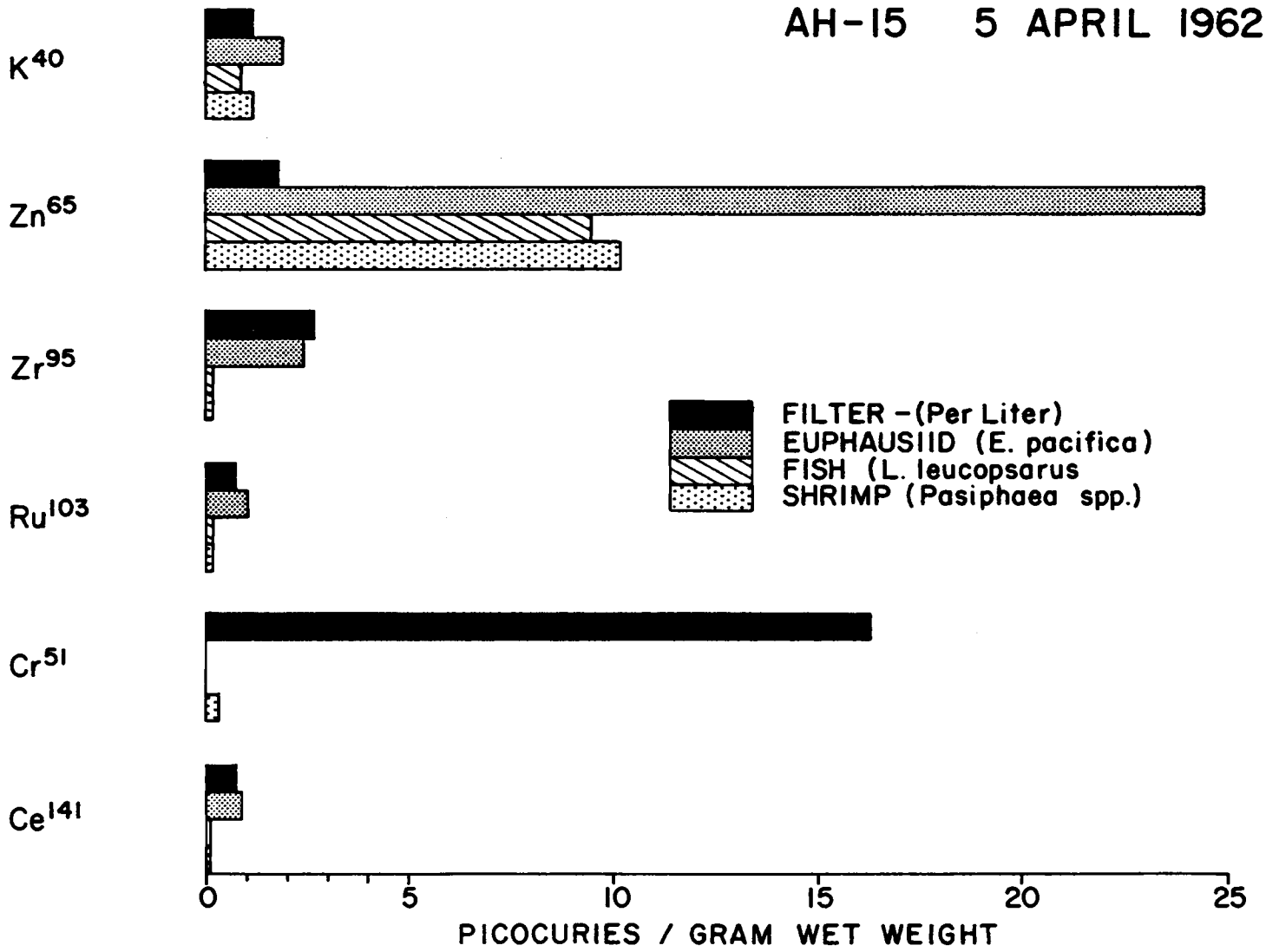

Figure 26. Comparison of radioactivity in three trophic levels

to these three fission products, chromium-51 was collected in quantity by the membrane filter, but not by the euphausiids. The reason for this is not clear. The form of chromium in sea water is not known (its valence state is CrO_4^-) (41, p. 5), but it is not particulate in fresh water, nor is it associated with particles (26). This must not hold true in sea water, as non-particulate chromium easily should pass through the membrane filter. Since the filter traps 90% of all material greater than 0.1 micron, an appreciable amount of the chromium must occur in or on particles larger than this. On the other hand, the particles must be too small to be removed from the water by the filtering setae of euphausiids. It seems unreasonable that this organism could select against all chromium associated with its food and still permit Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141} to enter its digestive tract, particularly since none of the four radionuclides seem to be biologically active.

The fifth element of interest is zinc-65. Zinc is ionic in sea water and ionic forms of radioactivity seem to be accumulated to a lesser extent by phytoplankton than do particulate forms (43, p. 135). Some marine organisms may take up ionic zinc directly from the water so that it need not appear in food particles in order to become incorporated in the tissues. For example, Boroughs et al (12) show that bay scallops immersed for 2 hours in sea water containing zinc-65 accumulated this isotope very rapidly. After only 3 hours, Zn^{65} was found

concentrated in the organs and tissues of the scallops, particularly the kidney. Euphausiids also probably assimilate ionic zinc, i.e. Zn^{++} or $ZnCl^{+}$, very rapidly. Yet some of the Zn^{65} must be associated with the particulate fraction of the sea water, because it was present on membrane filters.

Particulate zinc in sea water is a distinct possibility under certain conditions. In cases in which a nuclear blast has carried coral sand up into the fireball, normally soluble radiomaterials can be trapped inside particles (derived from the coral) that are insoluble in sea water (1, p. 45-54). However, these particular circumstances would not prevail in this case. A second possibility is somewhat more likely. Manganese hydroxide, $Mn(OH)_4$, has a weakly acid character and the hydrosol is therefore negatively charged (58, p. 649). Joyner (35, p. 20-21) suggests that it could therefore scavenge positive ions (such as zinc), just as ferric hydroxide is known to scavenge negative ions. However, the presence of zinc-65 in euphausiids taken as far as 250 miles from the mouth of the Columbia River seems to argue in favor of zinc being in solution in sea water. Ionic zinc-65 could be taken up directly from the water by euphausiids and phytoplankton and possibly adsorbed to detritus. This too would explain the observations.

The problem of the state of zinc-65 at the mouth of the Columbia River must be considered unresolved. Although zinc was retained on the filter, it is not known whether any radio-zinc remained in the filtrate. A somewhat puzzling addition is the recent evidence (3, p. 11) from the University of Washington which indicates that zinc-65 is one of the principal gamma emitters in sediments at fairly large distances from the mouth of the river. Apparently, biological and non-biological systems are competing for the zinc. The ocean, though grossly undersaturated with respect to this metal, continues to lose it to organisms and sediments.

Figure 27 shows an additional comparison of the radionuclides in euphausiids and lantern fish. The results are similar to those of Figure 26, except for zinc, which appears to be more concentrated in L. leucopsarus than in euphausiids. Similar results have been reported for tuna (69), whose specific activity of Zn^{65} was found to be greater than that of some of the small organisms on which it was presumed to feed. There is certainly no reason to believe that the concentration of Zn^{65} cannot increase at higher trophic levels, although under equilibrium conditions the greater total amount of zinc would be found associated with the greater biomass* of the lower trophic levels.

*To avoid arguments concerning "production" and "biomass," it is emphasized that this "biomass" need not be present at any one time but represents the biomass of successive generations of organisms from lower trophic levels.

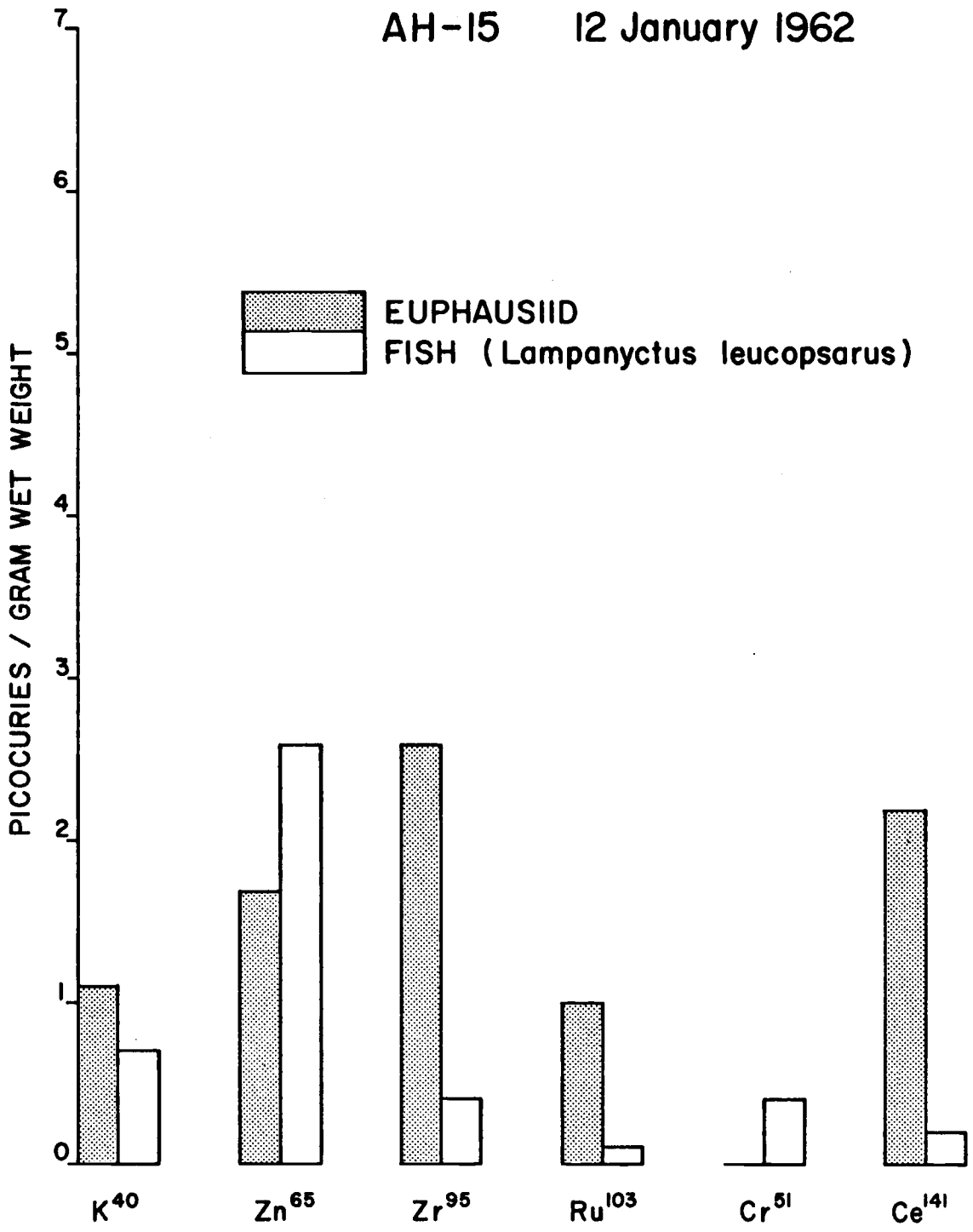

Figure 27. Comparison of radioactivity in euphausiid and lantern fish

In addition to euphausiids, salps, and lantern fish, another member of the macroplankton often found in some quantity was the sergestid shrimp, Sergestes similis. Figure 28 shows a comparison of the relative abundances of radionuclides in sergestids and euphausiids. Their food habits seem quite similar.

A series of hauls off Newport in August 1961 contained a few viper fish, Tactostoma macropus. Nine of these fish, from closely consecutive tows, were ashed and analyzed by gamma-ray spectrometry. The results, shown in Figure 29, indicate a lack of fission products prior to the Russian tests, but Zn^{65} again is present.

Radioanalyses cannot be substituted for basic ecological studies of the marine organisms. They may, however, show biological demand for certain elements that otherwise might be unsuspected. Certainly the importance of zinc in the marine environment must be amplified in view of its universal presence at all marine trophic levels investigated. Likewise, the almost total discrimination against chromium by macroplankton and nekton, in view of the amounts available in the water as evidenced by the membrane filter, is surprising. Cr^{51} is given as one of the important gamma emitters in net plankton off the mouth of the Columbia River (70, p. 7), also. Surprising, too, is the almost

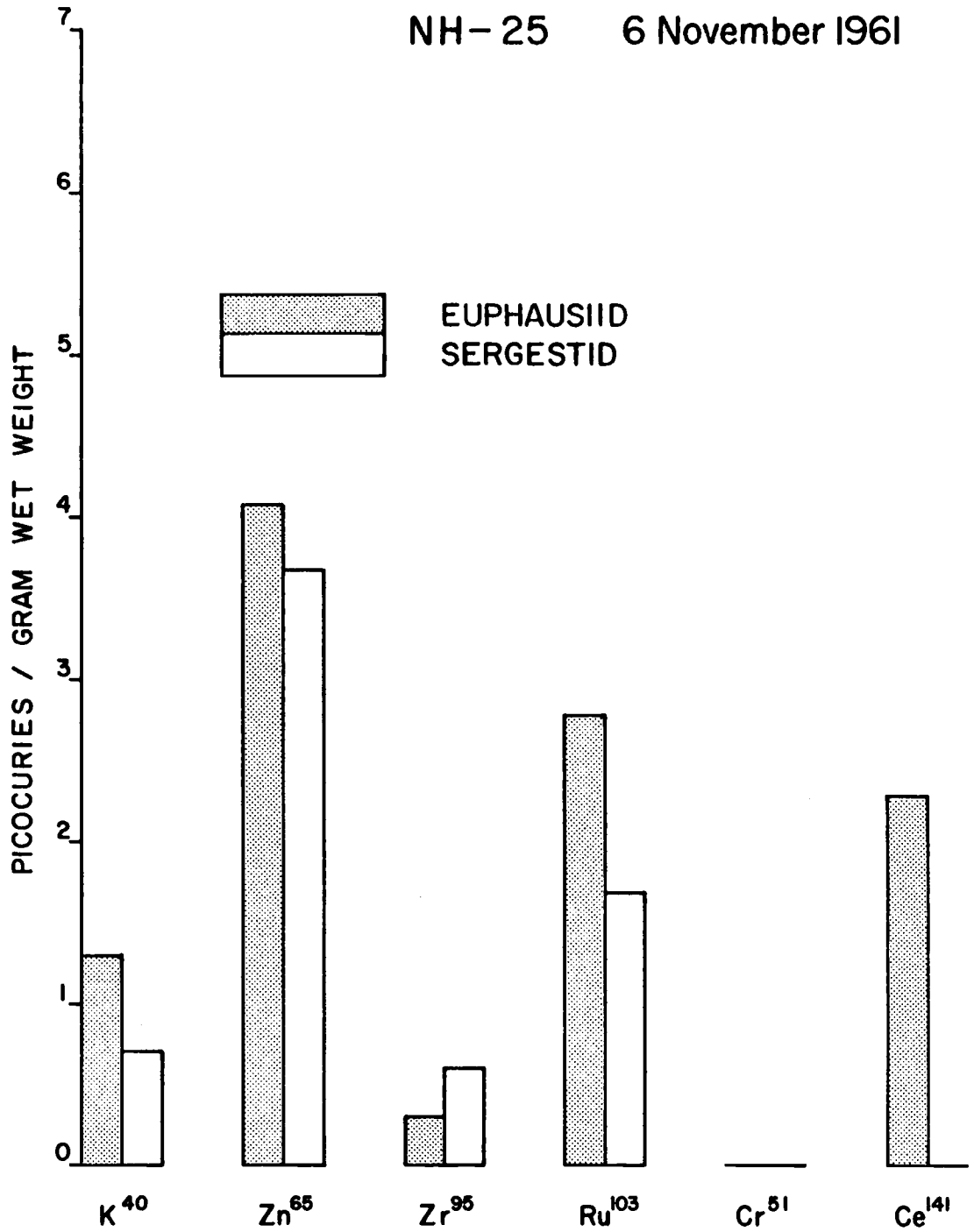

Figure 28. Comparison of radioactivity in euphausiids and sergestids

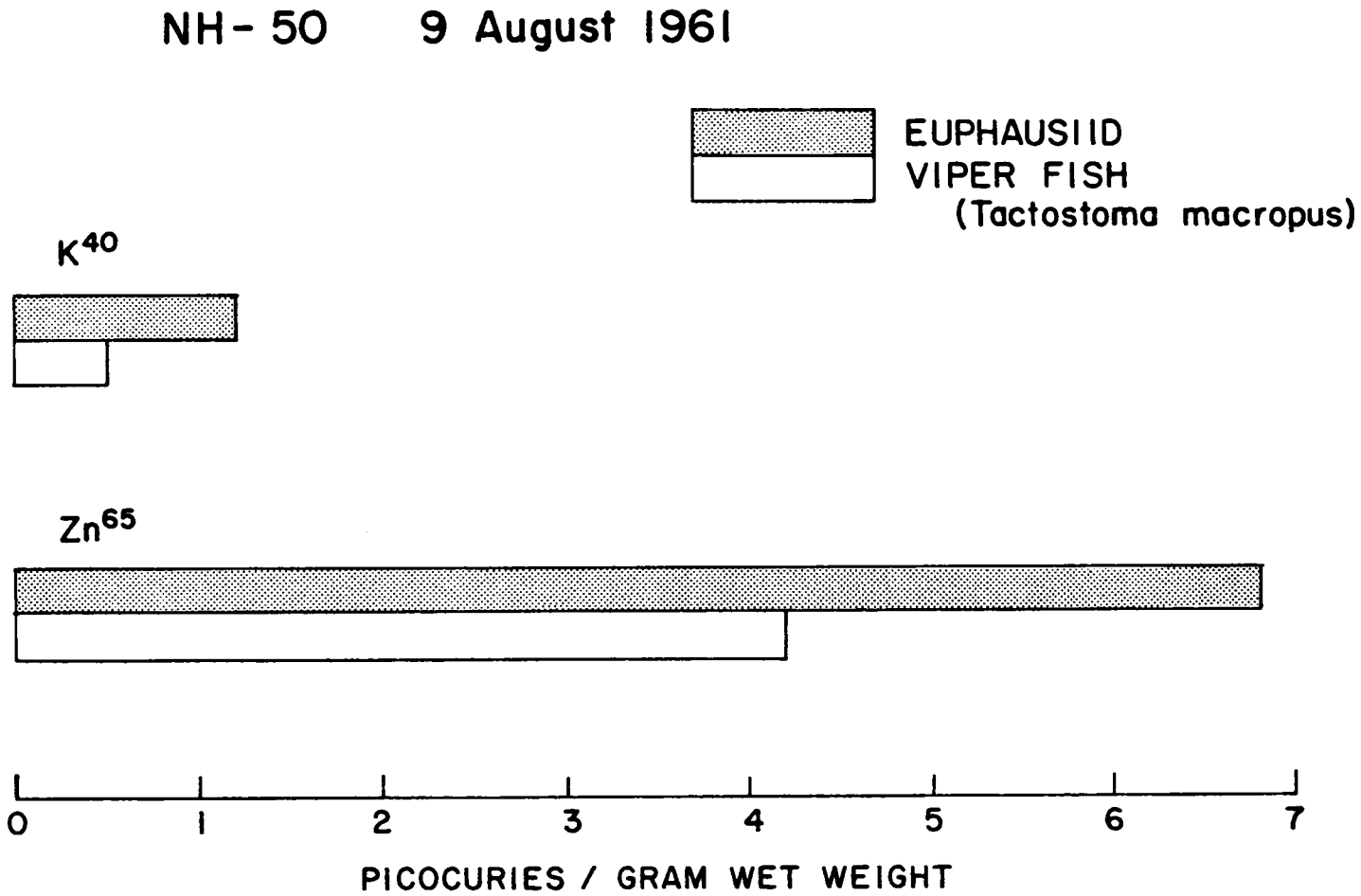

Figure 29. Comparison of radioactivity in euphausiids and viper fish

complete absence of Cs^{137}* and Co^{60} in the macroplankton and nekton tested herein. Co^{60} was listed as one of the principal gamma emitters in the plankton, according to a retesting of the TROLL survey samples (44). It also is found in rather large amounts in the sediments of the Columbia River (68), presumably there because it is derived from Hanford by neutron activation. Since it is a part of the structure of vitamin B_{12}, and like zinc it is a transition element, its absence from the organisms examined was unexpected. Cs^{137}, although not concentrated by plankton, is concentrated slowly to rather high levels in the muscles and soft tissues of shell fish and in fish (17). Its absence in the macroplankton must be largely due to the ionic form in which it exists in sea water (43, p. 117).

E. Variation in Replicate Tows

The question of variation in replicate tows is inherent in all biological sampling. Nevertheless, it was expected that radioanalysis of a sample consisting of hundreds of euphausiids (actually about 750 euphausiids per 13 cc sample) would agree rather closely with that of a

*A suggestion of Cs^{137} and Mn^{54} was found in the carid shrimp, Pasiphaea spp. taken 15 miles off Astoria, Oregon. Co^{60}, Cs^{137}, Mn^{54}, and Ru^{103} were found in an albacore tuna from the same area. Zinc-65 was the principal radioisotope in both of these organisms, however. Particulate fission products were negligible.

similar sample of more hundreds of euphausiids taken in the same location an hour later. This was not actually tested until later in the program because of the premium on counter time and the appearance of the data in the early work, which showed that zinc-65 activity did not vary greatly from station to station, except near the mouth of the Columbia River. This stability, which apparently was not characteristic of unsorted net plankton (63), gave a sense of confidence and indicated a certain degree of freedom from random errors.

Later in the study replicate tows, made through the night of July 17-18, 1961, were analyzed, utilizing both euphausiids and salps, and they tend to vary together, apparently responding to a true internal difference in the amount of radio-zinc (Figure 30). Despite the limited number of observations, there is the suggestion of a curve of radioactivity which lags behind the maximum feeding time (1 a.m.) by a couple of hours. The similarity of this apparent cycle to that of diurnal migration led to a perusal of the records for further replicate tows.

The more random results obtained in the replicate series of April 11-12, 1962 (Table 4), make the cycle seen in Figure 30 appear fortuitous. However, in view of the odds of obtaining merely by chance the effect noted, these observations should be repeated. Perhaps data from 2 successive nights would show a better correlation.

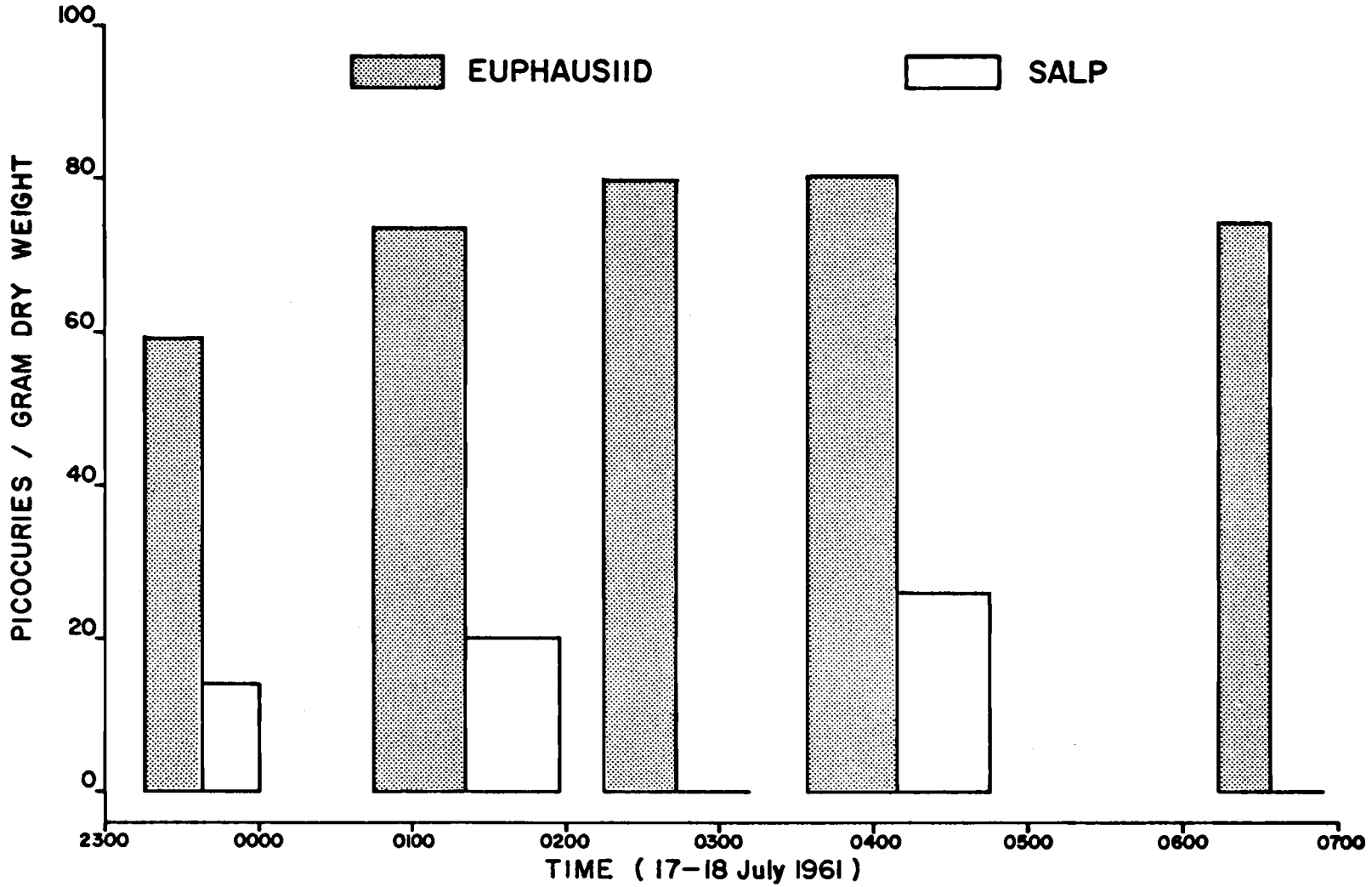

Figure 30. Zinc-65 changes in salps and euphausiids during a series of replicate tows, July 17-18, 1961

TABLE 4

Variability of Zn^{65}, Zr^{95}-Nb^{95}, Ru^{103} and Ce^{141} in Replicate Tows Made 50 Miles off Newport, Oregon April 11-12, 1962

	Zn^{65}	Zr^{95}-Nb^{95}	Ru^{103}	
MT				days elapsed between collection and counting
155*	14.2	3.2	4.4	114
157	10.7	15.0	6.2	114
158	8.5	14.3	5.7	"
159	11.3	11.3	5.2	"
160	9.6	14.2	9.1	"
161	10.4	15.2	8.4	"
162	11.5	13.2	3.9	"
163	11.2	13.9	5.9	"
164	14.5	13.4	6.9	"
165	11.0	12.4	3.7	"
Mean	11.0	13.7	6.1	
σ	1.5	1.1	1.7	

*MT 155 was not included in the calculations of the mean and σ.

The results of the radioanalyses of the replicate series of April 11-12 are shown in Table 4. Because of the presence of fallout fission products, data for zirconium-95 and ruthenium-103 are included with that of zinc-65. It shoud be noted that little scatter is observed in the values for the three isotopes during the course of the night. Ruthenium-103 is expected to show the greatest variation because of the low levels present (yielding poor statistics), and the errors inherent in this determination due to Compton continuum. The greatest source of error is because of the relatively short half-life of Ru^{103} (40 days) since 114 days elapsed between the time of collection and radioanalysis of these samples. This elapsed time increases the standard deviation by a factor of 7.2. The delay in counting is less serious for Zr^{95} both because of its longer half-life (65 days) and because it decays into Nb^{95}. The 245 day half-life of Zn^{65} should assure even better accuracy for it.

Perhaps the results for MT 155 (midwater trawl #155) are significant. It was the only daylight trawl in the series which yielded sufficient euphausiids for radioanalysis, and was also unique in that it was made at a greater depth (200-500 meters). The balance of the trawls were made during the hours of darkness at depths of from 0 to 200 meters. Note that zinc-65 and ruthenium-103 are essentially the same in MT 155 as they are in the night-time tows. Zirconium-95, however,

is considerably less. This may be due to the short retention time of Zr^{95}-Nb^{95}. The euphausiids in MT 155 were taken from beneath the zone of maximum phytoplankton and may have had empty digestive tracts. The tentative evidence presented herein suggests several applications which may be explored further and which may ultimately prove of value in marine ecological studies.

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APPENDICES

APPENDIX I

ORIGIN OF THE FOUR PRINCIPAL RADIOISOTOPES FROM HANFORD WHICH REACH THE OCEAN IN QUANTITY

The four principal radionuclides in the ocean as a result of the Hanford operations are formed principally through the radiative capture of thermal or epithermal neutrons passing through the reactors. The amount of each radioisotope in the river, and eventually in the ocean, thus ultimately depends on the neutron flux in the reactors, the amount of trace element (such as Zn^{64}) in the coolant river water, and the residence time of the coolant water in the reactors.

A. Phosphorus-32

This isotope originates from the following reaction:

$$_{15}P^{31} + {}_{0}n^{1} \longrightarrow {}_{15}P^{32} + \gamma$$

This process, radiative capture, is most important when the impinging neutron is thermal or epithermal, i.e. .035 to about 2 Mev. The nucleus of ordinary stable phosphorus can absorb a neutron and lose the excess energy of impact by the emission of a gamma photon. There is no Coulomb barrier against neutrons, and thermal neutrons are especially reactive. The resulting P^{32} possesses 17 neutrons

and 15 protons compared with the 16 neutrons and 15 protons of ordinary phosphorus.* Despite the loss of kinetic energy through the emission of a gamma photon in the capture process, the new nucleus formed is unstable because of the increased neutron to proton ratio, and decays by beta-emission:

$$_{15}P^{32} \xrightarrow{14.3 \text{ days}} {}_{16}S^{32} + \beta^- \ (E_{max} = 1.7 \text{ Mev.})$$

S^{32} is a stable isotope of sulphur.

B. Neptunium-239

Neptunium-239 originates from uranium-238 in a two-step process.

(1) $_{92}U^{238} + {}_{0}n^{1} \longrightarrow {}_{92}U^{239} + \gamma$

(2) $_{92}U^{239} \xrightarrow{23.5 \text{ min.}} \beta^- + {}_{93}Np^{239} + \gamma$

The radiation from Np^{239} is not final, however, because it decays to plutonium, etc., in the following series of reactions:

$$_{93}Np^{239} \xrightarrow{2.33 \text{ days}} \beta^- + {}_{94}Pu^{239}$$

$$_{94}Pu^{239} \xrightarrow{24,360 \text{ yrs}} \alpha + {}_{92}U^{235}$$

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* The stable isotope of phosphorus in nature is 100 percent P^{31}.

$$_{92}U^{235} \xrightarrow{7 \times 10^8 \text{ yrs.}} \alpha + {}_{90}Th^{231}$$

$$_{90}Th^{231} \xrightarrow{25.6 \text{ hrs.}} \beta^- + {}_{91}Pa^{231}$$

$$_{91}Pa^{231} \xrightarrow{3.4 \times 10^4 \text{yrs.}} \alpha + {}_{89}Ac^{227}$$

Here the series becomes complex since actinium-227 can decay by the emission of a negative beta particle (98.8%), or an alpha particle (1.2%). This whole sequence is called the actinium or 4N + 3 series (29, p. 13). It is a series of exoergic nuclear adjustments, terminating eventually in Pb^{207}, a stable isotope of lead. The heavy elements are not utilized by organisms, although they pose a hazard in some cases because they may be deposited in certain tissues. However, the levels expected in the ocean are extremely low, and the biggest problem is one of detection.

While the amount of Np^{239} in the Pacific Ocean as a consequence of Hanford is very low, it is nonetheless interesting to point out the buildup of daughter products of Np^{239}. Assuming a 10-day transit time from Hanford to Vancouver and using the figure of 72 curies per day past Vancouver, the amount of Np^{239} produced by Hanford would be:

$$N_o = Ne^{\lambda t}$$

$$= 72\, e^{2.97}$$

$$= \sim 1400 \text{ curies per day (at Hanford before any decay)}$$

The rate of Pu^{239} built up from the decay of Np^{239} would be:

$$\frac{dN_2}{dt} = N_1 \lambda_1 - N_2 \lambda_2$$

where N_1 is the amount of Np^{239}

N_2 is the amount of Pu^{239}

λ_1 is the decay rate of Np^{239} ($.297\ day^{-1}$)

λ_2 is the decay rate of Pu^{239} ($.78 \times 10^{-7}\ day^{-1}$)

Since λ_2 is practically zero, though, it can be ignored until N_2 becomes very large. It is approximately correct to say that at the end of 10 years (at the 1960 rate of production of Np^{239}), there would be produced:

$$1400 \text{ curies/day} \times 10 \text{ yrs} \times 365 \text{ days} = 5.1 \times 10^6 \text{ curies } Np^{239}$$

All of this would decay to Pu^{239}. Np^{239} has a short half-life (2.33 days), while Pu^{239} has a long half-life (2.436×10^4 years)(39, p. 97). Therefore since:

$$\frac{C_1}{C_2} \cdot \frac{(t\tfrac{1}{2})_2}{(t\tfrac{1}{2})_1}$$

then $$\frac{5.1 \times 10^6 \text{ curies } Np^{239}}{X \text{ curies } Pu^{239}} \times \frac{2,346 \times 10^4 \times 365 \text{ d.}}{2.33 \text{ d.}}$$

Solving for X gives 1.39 curies of Pu^{239} produced by Hanford (from the decay of Np^{239}) in ten years at the 1960 rate of production. While it is true that the amount of Pu^{239} that has accumulated in the ocean to date as a result of Hanford operations is probably less

than two curies, it is building up at a slow but steady rate (R), and, at some distant time, will reach an equilibrium value (Ne). This value can be computed from:

$$Ne = \frac{R}{\lambda} \qquad \text{where } \lambda = \frac{.693}{2.44 \times 10^4 \text{ yrs.}}$$

$$= 2.84 \times 10^{-5} \text{ yrs}^{-1}.$$

$$\text{and } R = .135 \text{ curies/yr.}$$

Then Ne would be 4.75×10^3 curies of Pu^{239} in the ocean if equilibrium were reached at the 1960 rate of production. It is prudent to point out, however, that after 24,360 years (the half-life of Pu^{239}), only some 2,380 curies will have accumulated, if the present rate is maintained. The buildup of Pu^{239} does not appear to be a problem of present concern.

C. Chromium-51

Chromium-51 is present in rather large amounts, even though Cr^{50}, its precursor, makes up only 4.31 percent of stable chromium. It is hard to explain the great quantities of Cr^{51} in the effluent unless the circumstances are considered. Sodium dichromate is added to the reactor coolant, i.e. the Columbia River, to inhibit corrosion in the pipes and coils of the system. Chromium, including Cr^{50}, is therefore present in greater than trace amounts. Also, Cr^{50} has a large nuclear cross section for thermal neutrons,

and thus many n,γ reactions are expected. The isotopes of chromium are (29, p. 405 and 418):

	Nuclear cross-section (barns)	% abundance	half-life
Cr^{50}	16.0	4.31	stable
Cr^{51}	RA	RA	27.8 days
Cr^{52}	0.73	83.76	stable
Cr^{53}	18.0	9.55	stable
Cr^{54}	0.3	2.38	stable
Cr^{55}	RA	RA	3.52 min.

Only Cr^{50} and Cr^{54} (becoming Cr^{51} and Cr^{55}, respectively) are activated by the neutron flux in the reactors. Cr^{54} plays a relatively small role in the radioactivity of the effluent because it has a small nuclear cross section, resulting in fewer nuclear transformations, it makes up only a small fraction of the chromium found in nature, and its radioactive product, Cr^{55}, has an extremely short half-life, decaying rapidly to stable manganese-55.

Despite the great apparent abundance of Cr^{51} in the ocean, it probably does not constitute much of a hazard to the biosphere because of its great dilution. Thirty three thousand curies of Cr^{51} (the computed equilibrium value in the ocean) would decay at the rate of 7.3×10^{16} disintegrations per minute, but this only equals

the radiation to be expected from about one-third of a gram of pure chromium-51. Cr^{51} has an activity mass relationship of 1.08×10^{-5} grams per curie, thus (39, p. 98):

$33,000 \text{ curies} \times 1.08 \times 10^{-5} = .358 \text{ grams of pure chromium-51}$

The mode of decay of Cr^{51} is by electron capture:

$$_{24}Cr^{51} \xrightarrow[\text{e. c.}]{27.8 \text{ days}} {}_{23}V^{51} + \gamma$$

D. Zinc-65

The last of the four radionuclides present in quantity in the ocean as a result of the activities of Hanford Laboratories is zinc-65. It is formed in the following reaction:

$$_{30}Zn^{64} + {}_{0}n^{1} \longrightarrow {}_{30}Zn^{65} + \gamma$$

Zinc-64 has a neutron cross section of 0.5 barns and makes up 48.89 percent of the zinc found in nature. The mode of decay is:

$$_{30}Zn^{65} \xrightarrow{245 \text{ days}} \beta^{+} + {}_{29}Cu^{65}$$

APPENDIX II

AMOUNT OF SALPS COLLECTED

MT	DATE	LOCATION	DEPTH	SALPS OUNCES
	June			
3	13VI 61	NH-50	135	1
4	"	"	200	1
5	no samples taken			
6	13VI61	NH-50	240	4
7	"	"	?	1
8	"	"	200	1 1/2
9	"	"	30	4 1/2
10	14VI61	"	200	6
11	"	"	"	2
12	"	"	"	6
13	"	"	"	5
14	"	"	"	7
15	"	"	500	15
	July			
16	17VII61	"	200	10
17*	"	"	"	8
18*	"	"	"	9
19	"	"	"	12
20	18VII61	"	"	16
21	"	"	"	5 1/2
22	"	"	"	13
23	"	"	"	6
24	"	"	500	2
25	18VII61	NH-50	1000	1/2
26	"	"	200	1/2
27	"	"	500	13
28	"	"	1000	4 1/2
29	"	"	500	8
30	"	"	1000	4
31	19VII61	"	500	2
		"	[illegible]	5
				[illegible]

MT	DATE	LOCATION	DEPTH	SALPS OUNCES
35	20VII61	AH-30	200	28
36	"	AH-20	"	4
	August			
37	1VIII61	CH-50	"	28
38	"	CH-40	"	56
39	2VIII61	CH-30	"	34
40	"	CH-20	"	7
41	9VIII61	NH-50	25	5 1/2
42	"	"	20	4 1/2
43	"	"	10	56
44	"	"	200	6
45	"	"	"	4
46	"	"	50	4
47	"	"	200	3
48	"	NH-40	"	2 1/2
49	10VIII61	NH-30	100	0
50	"	NH-20	75	0
51	"	NH-10	40	0

APPENDIX III

Sample Calculation

Raw Data (MT 144 E)

	Total Counts (-Bkg)	Bkg (Mean)	Total Counts (+ Bkg)
1. K^{40}	84	408	900
2. Zn^{65}	2367	410	3187
3. Zr^{95}	2726	319	3364
4. Ru^{103}	538	647	1832
5. Cr^{51}	384	1382	3148
6. Ce^{141}	754	779	2312

Total counting time was 30 minutes for MT 144 E(euphausiids) and 30 minutes for background, using 13 ml of distilled water as a blank.

1. K^{40} $\quad \frac{84}{30} = 2.8 \text{ cpm} \quad \pm \sqrt{\frac{900}{30}} = \pm 1.0$

$$K^{40} = 2.8 \text{ cpm} \pm 1.0 \text{ cpm}$$

2. Zn^{65} $\quad \frac{2367}{30} = 78.90 \pm \frac{\sqrt{3187}}{30} = \frac{56.5}{30} = \pm 1.88$

$$Zn^{65} \text{ apparent} = 78.90 \pm 1.88$$

But part of these counts are caused by Compton continuum from K^{40}. Table 2 shows that 4.4 percent of the counts recorded for K^{40} are present in the Compton continuum in the region of the zinc-65

photopeak.

K^{40} Compton = (2.8 ± 1.0)(0.0440) = 0.123 ± 0.44

Zn^{65} actual = Zn^{65} apparent - K^{40} Compton

Zn^{65} actual = 78.90 - 0.123 ± $\sqrt{(1.88)^2 + (.044)^2}$

= 78.78 ± 1.88

However the actual number of counts at time of counting is less than the number of counts at time of sample collection. In this case, the delay was 34 days. Using $N = N_o e^{-\lambda t}$ where $\lambda = \frac{0.693}{t}$, and t = 245 days, we obtain a ratio for $\frac{N}{N_o} = 1.101$

1.101 (78.78 ± 1.88) = 86.74 ± 2.07, which is the actual number of counts to be expected at the time the sample was collected.

3. Zr^{95} $\frac{2726}{30} = 98.87 \pm \frac{\sqrt{3364}}{30} = \frac{58}{30} = 1.933$

Zr^{95} app. = 98.87 ± 1.933

Compton correction (from Table 2)

K^{40} = (2.8 ± 1.0)(0.375) = 0.1050 ± 0.0375

Zn^{65} = (78.78 ± 1.88)(0.0459) = 3.616 ± 0.0863

Zr^{95} act. = (98.87 - 0.105 - 3.616) ± $\sqrt{(1.933)^2 + (.0375)^2 + (.0863)^2}$

= 87.146 ± 1.94

Half life correction ($e^{\lambda t}$) = 1.438

1.438 (87.146 ± 1.94) = 125.316 ± 2.790

4. The same procedure is followed for Ru^{103}, Cr^{51}, and Ce^{141}

using appropriate constants (for Compton correction) from Table 2.

5. After all 6 isotopes have been corrected for Compton continuum and decay, counts per minute per total sample must be converted to picocuries per gram. The final corrected counts must be:

(a) multiplied by efficiency factor (Table 2)

(b) divided by sample size in grams

(c) divided by 2.22 (constant used to convert counts per minute to picocuries)

$K^{40} = 2.8 \pm 1.0$ Efficiency factor = 40.55

$\frac{2.8 \times 40.55}{6.82 \times 2.22} = 7.5$ Sample size = 6.82

$\frac{1.0 \times 40.55}{6.82 \times 2.22} = 2.68$ $K^{40} = 7.5 \pm 2.7$ picocuries

$Zn^{65} = 86.74 \pm 2.07$ Efficiency factor = 6.00

$\frac{86.74 \times 6.00}{6.82 \times 2.22} = 34.4$ Sample size = 6.82

$\frac{2.07 \times 6.00}{6.82 \times 2.22} = 0.82$

$Zn^{65} = 34.4 \pm 0.8$ picocuries

Zr^{95}, Ru^{103}, Cr^{51}, and Ce^{141} are treated in the same manner, using the efficiency factors listed in Table 2.

APPENDIX IV

ABUNDANCE OF EUPHAUSIIDS

All midwater trawls from 1 through 234 were examined, with the exception of trawls 179 through 187. This last group was a special series made off Brookings, Oregon and was not a part of the routine program. Several other trawls of a special nature were not used, principally because there were tows of short duration for demonstration purposes and were non-routine. Excluding these, a total of 220 trawls were inspected to see if euphausiids were present in sufficient quantity for radioanalysis. The first tow was made March 28, 1961. Data are complete through Sept. 7, 1962.

There were 5 categories used to describe the quantities of euphausiids present:

1. NONE — If no euphausiids were present.
2. FEW — If some were present, but obviously not enough for radio analysis.
3. PERHAPS — Probably enough for radioanalysis, but some uncertainity.
4. ENOUGH — Obviously enough (50 g.) but not a pint.
5. MANY — More than a pint.

A break down of the relative abundance of euphausiids is given below:

AMOUNT	TOTAL NUMBER	%
None	16	7.3
Few	36	16.4
Perhaps	26	11.8
Enough	46	20.9
Many	96	43.6
Total	220	100.0

DAYLIGHT TOWS

None	8	22.2
Few	13	36.1
Perhaps	5	13.9
Enough	6	16.7
Many	4	11.1
	36	100.0

NIGHTTIME TOWS

None	8	4.4
Few	23	12.5
Perhaps	21	11.4
Enough	40	21.7
Many	92	50.0
	184	100.0

It is apparent that nighttime tows (normally of 30 minutes duration) usually supply sufficient euphausiids for radioanalysis. Only about 17% of these tows fail to provide sufficient euphausiids for radioanalysis. The deeper daytime tows generally were more productive than surface tows.